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INTERNAL FRICTION IN IRON AT LOW TEMPERATURES

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As part of an extensive investigation of dislocation relaxation phenomena in fcc and bcc metals, internal friction measurements have been made on annealed and cold worked iron. Results on other metals including copper, aluminum, and niobium will be published soon.

Measurements have been made over the temperature range 4.2°K to 350°K using iron bars driven in a longitudinal mode of vibration at a resonant frequency of about 25 kc/sec. An electrostatic drive is employed; internal friction is measured by a free decay method.

The data on iron are summarized in Fig. 1; a typical loss curve for cold-worked copper is included for comparison. It will be noted that the results for copper are similar to those initially obtained by Bordoni[1] and later by several other workers.[2-4] It appears that in iron the dislocation relaxation peaks characteristic of fcc metals are absent or at least very much attenuated. Preliminary results on another bcc metal, niobium, indicate a similar absence of such peaks.

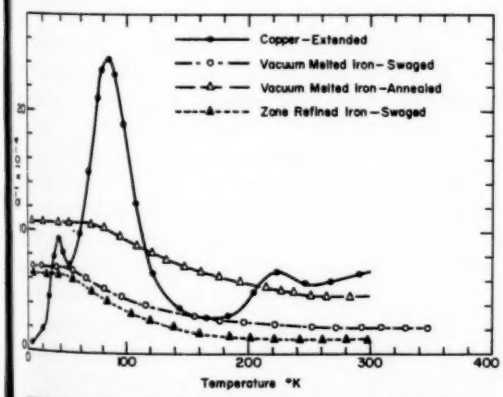

FIG. 1. Internal friction of iron and copper as a function of temperature.

The results on iron are atypical in that the internal friction increases with decreasing temperature, reaching a maximum at around 50°K and maintaining that value to the lowest temperature reached. This behavior is not understood but may be related to the fact that the material is ferromagnetic.

The data presented in Fig. 1 summarize results obtained on two iron specimens. One was prepared from vacuum-melted material, the other from iron which had been zone-refined. The aim was to use material which was as free of interstitial impurities as possible. The results of analysis are given in the following table.

Specimen	O (ppm)	N (ppm)	H (ppm)	C (ppm)
Vacuum-melted iron	9	37		18
Zone-refined iron	17	<2	<0.2	10

The specimen of vacuum-melted iron was reduced in area 8% by swaging prior to the initial measurements. It was remeasured after a four-hour vacuum anneal at 850°C. The specimen of zone-refined material was rolled and swaged from an ingot measuring $1\frac{3}{4}$ in. $\times 1\frac{1}{4}$ in. $\times \frac{3}{4}$ in. to a final diameter of $\frac{3}{8}$ in. to give a total reduction of 90%. No intermediate anneals were required.

The mechanism of dislocation relaxation proposed by Seeger[5] does not seem to offer an explanation for the apparent absence of peaks in bcc metals. A principal difference between dislocations in fcc and bcc metals is that for the latter a dissociation of complete dislocations into symmetric partials is not possible. The insensitivity of the peaks to impurity content indicates that intrinsic point defects such as vacancies may be important. These facts suggest a new mechanism based upon the thermally activated motion of symmetric paired partial dislocations between vacancy-pinning points; minimum energy configurations are those for which the vacancy is immediately above either partial. It is in these positions that its interaction with the dilatational stress field of the partial will be strongest. The activation energy is determined by the energy required for the transition (by glide of the dislocation pair) between the two stable states. This model will be discussed in detail in a forthcoming publication.[6]

We wish to acknowledge the support and encouragement of Professor A. W. Lawson throughout the course of this work. The zone-refined iron was obtained through the courtesy of J. W. Halley, K. K. Fetters, and G. W. Rengstorff; it was purified by the latter under the project supported by the American Iron and Steel Institute.

[1]P. G. Bordoni, J. Acoust. Soc. Am. 26, 495 (1954).
[2]D. H. Niblett and J. Wilks, Phil. Mag. 2, 1427 (1957).
[3]H. L. Caswell, J. Appl. Phys. 29, 1210 (1958).
[4]D. O. Thompson and D. K. Holmes, J. Appl. Phys. 30, 525 (1959).
[5]A. Seeger, Phil. Mag. 1, 651 (1956).
[6]L. J. Bruner (to be published).

CRYSTAL STABILITY AND THE THEORY OF FERROELECTRICITY

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The condition that a crystal should be stable for all small deformations is that all the normal modes should have real frequencies.[1] The limit of stability against a particular mode of vibration is approached as the corresponding frequency approaches zero. In what follows we show that there is reason to believe that a ferroelectric transition, at least in certain crystals, is associated with such an instability or near-instability. The condition for a ferroelectric transition is therefore a problem in lattice dynamics, and when it is treated as such new insight into the problem is obtained. We illustrate this by means of a simple example, in which it is shown that it is possible for a diatomic cubic crystal to exhibit properties which are remarkably similar to those of barium titanate.

A shell model for the ions in an alkali halide crystal, proposed by Dick and Overhauser,[2] has been found to give a satisfactory explanation of the dielectric properties of alkali halides[3,4] and of the lattice dynamics of sodium iodide.[5] A similar model accounts quite well for the observed relation between frequency ω ($=2\pi\nu$) and wave vector $\vec{q}$ ($q=2\pi/\lambda$) of certain of the normal modes of germanium.[6] The theory of Woods, Cochran, and Brockhouse,[5] based on this model, gives the following expressions for the frequencies of the transverse optic (T.O.) and longitudinal optic (L.O.) modes of wave vector zero in a diatomic cubic crystal:

$$\mu\omega_T^2 = R_0' - 4\pi(\epsilon+2)(Z'e)^2/9v, \tag{1}$$

$$\mu\omega_L^2 = R_0' + 8\pi(\epsilon+2)(Z'e)^2/9v\epsilon. \tag{2}$$

Here μ is the reduced mass of the ions, ϵ the high-frequency dielectric constant (the square of the optical refractive index), and v the volume of the unit cell. $Z'e$ is the effective ionic charge, while $R_0'\vec{u}$ is the restoring "short-range" force on any one atom when the two lattices carrying the nuclei of different type are displaced a small relative distance $\vec{u}$. Primed quantities depend explicitly on the parameters of the shell model. In a crystal such as sodium iodide the two quantities on the right of Eq. (1) are of the same order of magnitude, but the term R_0' which arises from the short-range interaction is about twice as great as the other, which arises from the Coulomb interaction. Let us consider the

situation that would arise if their difference approached zero.

It is found from the theory[5] that ω_T may approach zero without the crystal necessarily becoming unstable against other vibration modes; indeed all transverse optic modes for which $\vec{q}$ is not close to zero may retain quite usual frequencies. The quantities which appear on the right of Eq. (1) may be temperature dependent, as the lattice vibrations are in practice not completely harmonic, so that near $T=T_c$ we may postulate

$$\frac{\mu\omega_T^2}{R_0'}=1-\frac{4\pi(\epsilon+2)(Z'e)^2}{9vR_0'}=\gamma(T-T_c), \qquad (3)$$

where γ is a temperature coefficient and T_c is the temperature at which the crystal would become unstable. It has been shown by Lyddane, Sachs, and Teller[7] that

$$\omega_L^2/\omega_T^2=\epsilon_0/\epsilon, \qquad (4)$$

where ϵ_0 is the static dielectric constant. Combining these equations, one finds

$$\frac{\epsilon_0-1}{4\pi}\simeq\frac{\epsilon_0-\epsilon}{4\pi}=\frac{(\epsilon+2)^2(Z'e)^2}{9vR_0'\gamma(T-T_c)}, \qquad (5)$$

so that a Curie-Weiss law is followed with a Curie constant

$$C=(\epsilon+2)^2(Z'e)^2/9vR_0'\gamma\simeq(\epsilon+2)/4\pi\gamma. \qquad (6)$$

The condition $\epsilon_0\to\infty$ is thus $\omega_T^2\to 0$. The ionic polarizability of one unit cell is found to be given by

$$\alpha_i=(Z'e)^2/R_0', \qquad (7)$$

while the electronic polarizability is as usual given by

$$4\pi\alpha_e/3v=(\epsilon-1)/(\epsilon+2). \qquad (8)$$

The condition $\omega_T^2=0$ is therefore the same as

$$(4\pi/3v)(\alpha_i+\alpha_e)=1, \qquad (9)$$

so that the terms "instability" and "polarizability catastrophe" are synonymous in this instance.

If one now postulates that the short-range potential between the two Bravais lattices is neither precisely harmonic nor precisely isotropic for comparatively large displacements, it can be shown that the crystal may become spontaneously polarized without becoming completely unstable. Instead it makes a transition to another phase. When the short-range potential is given explicitly by

$$V_R=\tfrac{1}{2}R_0'(u_x^2+u_y^2+u_z^2)+\tfrac{1}{4}B(u_x^4+u_y^4+u_z^4)+\tfrac{1}{6}B'(u_x^6+u_y^6+u_z^6)+\tfrac{1}{2}B''(u_x^2u_y^2+u_y^2u_z^2+u_z^2u_x^2), \qquad (10)$$

with $B<0$, B' and $B''>0$, the crystal makes a first-order transition to a tetragonal phase at a temperature exceeding T_c. Equation (10) refers to the "free" crystal; the potential V_R cannot therefore be used as it stands to give ω_T in a noncubic phase, since, for the lattice vibrations, the crystal is "clamped" by its inertia. At the cubic-tetragonal transition the atoms become displaced a relative amount

$$u_0=(3|B|/4B')^{1/2},$$

along a crystallographic axis, say [001]. The minimum T.O. frequency is reached just before the transition, and is given by

$$\mu(\omega_T^2)_{\min}=3B^2/16B'.$$

Expressions for the spontaneous polarization as a function of temperature, etc., are also obtained in terms of the above atomic parameters. As the temperature decreases still further the crystal makes a second transition in which the atoms become relatively displaced along [011], followed by a third which leaves them displaced along [111]. These results will be understood when it is pointed out that Eqs. (3) and (10) lead eventually to an expression for the free energy G_1 of the unstressed crystal which is almost identical with that postulated by Devonshire.[8]

The theory has been extended to apply to antiferroelectric transitions in diatomic crystals, and to ferroelectric transitions in other cubic crystals, including barium titanate. The equations which apply to the latter are greatly simplified by assuming that in the T.O. mode of lowest frequency (there are three T.O. modes in all), or in a static field, the framework of oxygen atoms is not distorted. The crystal structure analyses[9,10] of tetragonal barium titanate and lead titanate support this assumption. The dielectric properties of barium titanate, and the movements of barium and titanium atoms relative to the oxygen octahedron, may be accounted for by assuming a temperature dependence of certain atomic parameters analogous to that given by Eq. (3), and a short-range po-

tential for relative movement of titanium and oxygen atoms given by Eq. (10). A very small departure from a harmonic potential is found to be sufficient to account for the dielectric properties, and the numerical values required for other atomic parameters are physically reasonable.

The equation corresponding to Eq. (4), applicable to a perovskite-type crystal, is found to be

$$(\omega_2\omega_3\omega_4)_L^2/(\omega_2\omega_3\omega_4)_T^2=\epsilon_0/\epsilon. \qquad (11)$$

(This result does not depend on the assumption of an undistorted oxygen framework.) The infrared absorption frequencies $(\omega_3)_T$ and $(\omega_4)_T$ have been measured by Last,[11] and are in no way unusual. It follows from the theory that $(\omega_2)_T^2$ should be proportional to $(T-T_C)$ in the cubic phase, and should reach an abnormally low value estimated as $\nu=2$ or 3×10^{11} cps just before the first transition. This frequency should split appreciably in the tetragonal phase, and each frequency in this phase should vary inversely as the square root of the corresponding "clamped" dielectric constant. Although relaxation of the dielectric constant of barium titanate has been reported for frequencies of the order 10^{10} cps, Benedict and Durand[12] found that when a single crystal is used, the dielectric constant of the cubic phase is the same as the static value up to $\nu=2.4\times10^{10}$ cps, the limit of their experiment. The resonance frequency predicted here lies in the millimeter wavelength range. A study of the properties of barium titanate in this difficult frequency range should yield interesting results.

The statement that the problem of the onset of ferroelectric properties in any crystal is a problem in lattice dynamics is probably correct, but at present there is little prospect of detailed application of the theory of lattice dynamics to low-symmetry or disordered crystals.

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[1]M. Born and K. Huang, Dynamical Theory of Crystal Lattices (Oxford University Press, Oxford, 1954).
[2]B. G. Dick and A. W. Overhauser, Phys. Rev. 112, 90 (1958).
[3]J. E. Hanlon and A. W. Lawson, Phys. Rev. 113, 472 (1959).
[4]W. Cochran, Phil. Mag. (to be published).
[5]Woods, Cochran, and Brockhouse, Bull. Am. Phys. Soc. 4, 246 (1959).
[6]W. Cochran, Phys. Rev. Letters 2, 495 (1959).
[7]Lyddane, Sachs, and Teller, Phys. Rev. 59, 673 (1941).
[8]A. F. Devonshire, Advances in Physics, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1954), Vol. 3, p. 85.
[9]Shirane, Danner, and Pepinsky, Phys. Rev. 105, 856 (1957).
[10]Shirane, Pepinsky, and Frazer, Phys. Rev. 97, 1179 (1955).
[11]J. T. Last, Phys. Rev. 105, 1740 (1957).
[12]T. S. Benedict and J. L. Durand, Phys. Rev. 109, 1091 (1958).

NEW MECHANISM OF ANTIFERROMAGNETISM

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The well-known anomalous properties of dilute Cu-Mn alloys suggest an antiferromagnetic mechanism that requires neither two-body interactions between paramagnetic solute spins, nor a sublattice structure. Such a mechanism may be recognized by considering a new-type excited state of the conduction electron gas. This state will have the following properties. The expectation value $\hbar\vec{s}$ of the total conduction electron spin density operator, relative to an axis of quantization $\vec{\epsilon}$, will vary with position $\vec{R}$ as follows:

$$\vec{s}=bN\vec{\epsilon}\cos(\vec{q}\cdot\vec{R}), \qquad (1)$$

where b is a dimensionless amplitude and N is the number of atoms per cc. This static spin density wave will allow the total charge density of the gas to remain spatially uniform. Such a collective excited state may be visualized as the sum of two charge density waves, one of electrons with spin parallel to $\vec{\epsilon}$ and the other of antiparallel spin, which are equal in magnitude but opposite in phase. This state must be dynamically self sustaining, which is to say that it should be an eigenfunction of the Hartree-Fock equations. The possibility of such a self-con-

sistent solution results from the attractive character of the exchange potential operator A:

$$A\psi(\vec{r}_1) \equiv -\sum_{\vec{k}}[\int \phi^{\dagger}_{\vec{k}}(\vec{r}_2)\psi(\vec{r}_2)V(r_{12})d^3r_1]\phi_{\vec{k}}(\vec{r}_1), \quad (2)$$

where the set of (two component) functions $\phi_{\vec{k}}(\vec{r})$ are the occupied one-particle states of the electron gas, to be determined in a self-consistent way.

Define the following matrix elements of A between the space parts of, say, spin-up plane waves:

$$g^{\pm}(\vec{k}) \equiv \left(e^{i(\vec{k}\pm\vec{q})\cdot\vec{r}}\,|A|\,e^{i\vec{k}\cdot\vec{r}}\right). \quad (3)$$

These matrix elements will be small, proportional to the amplitude of the spin density wave, so that the perturbed plane wave (spin-up space) states of the electron gas may be written

$$\phi_{\vec{k}} = e^{i\vec{k}\cdot\vec{r}} + (g^{+}/\Delta^{+})e^{i(\vec{k}+\vec{q})\cdot\vec{r}} + (g^{-}/\Delta^{-})e^{i(\vec{k}-\vec{q})\cdot\vec{r}}, \quad (4)$$

where

$$\Delta^{\pm}(\vec{k}) = E(\vec{k}) - E(\vec{k}\pm\vec{q}).$$

If the functions (4) are inserted into (2) and the matrix elements (3) evaluated explicitly, one obtains the following pair of coupled integral equations for $g^{\pm}(\vec{K})$:

$$g^{+}(\vec{K}) = -\sum_{\vec{k}}\{[J(\vec{K}-\vec{k})g^{+}(\vec{k})/\Delta^{+}] + J(\vec{K}-\vec{k}+\vec{q})g^{-}(\vec{k})^{*}/\Delta^{-}\},$$
$$g^{-}(\vec{K}) = -\sum_{\vec{k}}\{[J(\vec{K}-\vec{k}-\vec{q})g^{+}(\vec{k})^{*}/\Delta^{+}] + J(\vec{K}-\vec{k})g^{-}(\vec{k})/\Delta^{-}\}, \quad (5)$$

where $J(\vec{q})$ is the Fourier transform of $V(\vec{r})$. By a not very straightforward procedure one can show that all the solutions of (5) can be constructed from solutions of two uncoupled integral equations:

$$h(\vec{K}) = -\sum_{\vec{k}}[J(\vec{K}-\vec{k}) \pm J(\vec{K}+\vec{k}+\vec{q})]h(\vec{k})/\Delta^{+}, \quad (6)$$

each solution of (6) generating two degenerate linearly independent solutions of (5), so as to form a basis which allows the phase of the spin density wave (1) to be varied continuously. For example, the two solutions of (5) corresponding to a solution $h(\vec{K})$ of (6), with the + sign, may be taken to be

$$g^{+}(\vec{K}) = h(\vec{K}), \quad g^{-}(\vec{K}) = h(-\vec{K})^{*};$$
$$g^{+}(\vec{K}) = ih(\vec{K}), \quad g^{-}(\vec{K}) = -ih(-\vec{K})^{*}.$$

The wave functions (4) for spin-up and spin-down electrons will differ only in the sign of $g^{\pm}$.

The wave vector $\vec{q}$ of the spin density wave plays the role of an eigenvalue parameter in the integral equations. If one neglects the variation of $J(\vec{K})$, the integral equations (6) have but one nontrivial solution, the magnitude of $\vec{q}$ being uniquely determined by

$$1 = (4J'/3\xi E_F)p(q/2k_0), \quad (7)$$

where J' is the average exchange energy per electron of the Fermi gas with Fermi energy E_F and Fermi wave number k_0, and where

$$p(x) = \tfrac{1}{2} + [(1-x^2)/4x]\ln[(1+x)/|1-x|],$$

a function which is unity at 0 and decreases monotonically with increasing x. $\xi(q)$ is a function, of unit order of magnitude, which relates the appropriate average of $J(\vec{K})$ to J' and which can be evaluated only by solving (6) exactly. Since $J' \sim E_F$ for most metals, it seems likely that Eq. (7) has roots for some metals, so that a static spin density wave is possible. The approximate solution is

$$g^{+} = g^{-} = -8J'b/9\xi = \text{constant}. \quad (8)$$

The energy (per cc) of the spin density wave is proportional to the square of its amplitude, and is

$$W = 8NCb^2/9\xi, \quad (9)$$

where C is the correlation energy per electron of the Fermi gas. This correlation energy (~1 ev/atom) appears alone in (9) because the uncorrected Hartree-Fock energy of the spin density wave is 0 (to order b^2). It should be appreciated that q, being determined by (7), will generally be incommensurate with reciprocal lattice vectors.

Paramagnetic solute spins will interact with a spin density wave through the ordinary s-d exchange interaction. A solute spin $\vec{S}_j$ at lattice site $\vec{R}_j$ will experience an effective field defined by the interaction Hamiltonian,

$$H_j = -Gb\vec{S}_j\cdot\vec{\epsilon}\cos(\vec{q}\cdot\vec{R}_j), \quad (10)$$

where G, as estimated from spectroscopic data, may be about 0.5 ev. Below a critical temperature T_c the orientation of solute spins by their interaction (10) allows the interaction energy to more than compensate the increased energy (9) associated with the spin density wave. Therefore, the amplitude of the wave becomes nonzero and long-range order of the solute spins

occurs, even though they are randomly distributed. A given solute spin will be polarized up or down depending on its location relative to the phase of the spin density wave, which necessarily has long-range coherence. Since the wavelength of the wave is incommensurate with the lattice parameter, the solute spins will experience a distribution in molecular fields B varying from $-Gb$ to Gb according to the probability distribution

$$P(B) = (G^2b^2 - B^2)^{-1/2}/\pi. \tag{11}$$

The thermodynamic properties of such a system may be derived in a straightforward manner. One finds

$$kT_c = 3\xi S(S+1)G^2c/32C, \tag{12}$$

where c is the solute concentration. This result is in excellent agreement with experiment. The low-temperature magnetic specific heat C_M is proportional to temperature:

$$C_M = \gamma_M T, \tag{13}$$

where

$$\gamma_M = 32\pi^2NCk^2/27G^2(2S+1)\xi. \tag{14}$$

The outstanding feature is that γ_M is independent of solute concentration. Recent experiments of J. E. Zimmerman (to be published) give remarkable confirmation of this result for Cu-Mn alloys having solute concentrations from 1/6 to 4 atomic percent. This linear specific heat is a direct consequence of the distribution (11). The reason is that each incremental increase in T, from 0°K, allows a new group of solute spins to reach the temperature at which they become thermally disoriented, and the integrated heat capacity of such a group is proportional to the temperature at which disorientation occurs. The lack of concentration dependence results from the fact that the spin density wave amplitude is proportional to c, so that $cNP(0)$ is independent of c. The magnetic specific heat is approximately linear in T all the way to T_c, at which point it drops to 0. T_c is about 25°K per atomic percent.

The anomalous electric, magnetic, and resonance properties of Cu-Mn alloys can be understood within the framework of this theory. They will be elaborated in detail in future publications.

RESISTANCE MINIMUM AND RESISTIVITY OF COPPER AT LOW TEMPERATURES

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The existence of an electrical resistance minimum in copper has been ascribed to the presence of small concentrations of specific metallic solutes in the metal by some workers,[1] while others have suggested structural defects in the metal as playing a major role.[2] Recently Redman et al. had demonstrated that the resistivity of single crystals of copper is markedly dependent on their thermal history.[3] In particular, the resistivity at the normal boiling point of He was found to decrease with increasing exposure time in an annealing treatment at 700°C in 30 μ of air and would attain a constant value after five hours of heat treatment. A re-examination of the earlier work on the resistance minimum in copper[2] in light of these new data suggested the possibility that the resistance minimum was influenced by the hardness of the vacuum during the annealing treatment. MacDonald and Pearson[1] also observed that the anomalous resistivity minimum seemed strongly dependent on reducing or oxidizing atmospheres during the melting of copper. A study of the role of the annealing atmosphere on the electrical resistivity minimum of copper was therefore felt to be of considerable interest.

The experimental techniques for these measurements are sketched briefly here with a detailed description reserved for later publication. The samples consisted of 0.05-cm diameter copper wire of 25-cm length wound around a quartz tube. The specimens were annealed in an evacuated mullite tube which allowed for annealing in desired atmospheres by means of a controlled leak. The resistance measurements were made on a Wenner potentiometer. Provisions for pumping on the He bath and a heater coil around the thermometer bulb which also served as the sample container enabled a temperature range to be

covered from 1.7°K to 25.0°K.

One particular sample of copper was subjected to eleven heat treatments in alternate oxidizing and reducing atmospheres. In every case annealing in a reducing atmosphere increased the overall resistivity and brought on a resistance minimum. Annealing in an oxidizing atmosphere reduced the resistivity and washed out the minimum. Curve 1 of Fig. 1, which is typical after a reducing treatment, was obtained by annealing the sample in a graphite boat in a vacuum of 2×10^{-5} mm Hg. A resistance minimum is found at about 12.5°K and an abrupt increase in the resistivity is seen in the region from 6°K to 8°K. The sharp rise was found to be due to the method of attaching electrical contacts to the sample and is incidental to the appearance of the resistance minimum. A typical curve following an air anneal (10 microns at 750°C for 2 hours) is shown in Curve 2 of Fig. 1. The resistivity at 4.2°K has decreased by approximately a factor of five and the minimum is not observable. An activation analysis revealed the presence of iron as the major impurity and smaller amounts of silver and zinc. Unfortunately it was not possible to obtain numerical values for these concentrations.

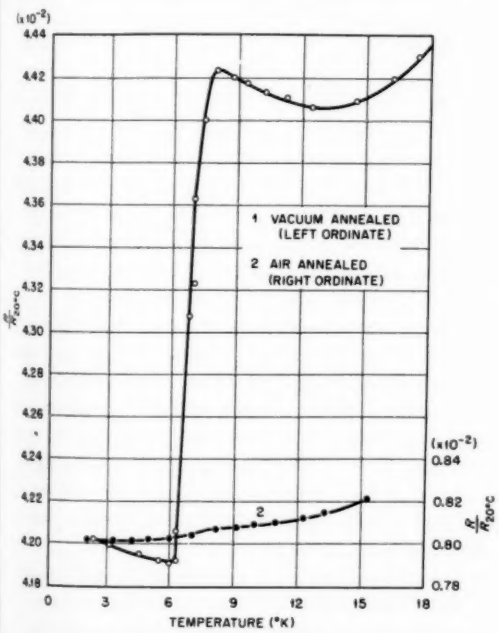

FIG. 1. The temperature dependence of the resistivity of copper with a small impurity content after annealing in a reducing atmosphere and an oxidizing atmosphere. (The sharp rise in resistivity was found to be due to the method of attaching electrical leads. Spot-welded leads removed this rise but did not affect the curves elsewhere.)

Similar results were found in a copper sample of much higher purity. In this case the specimen was annealed in a vacuum of 2×10^{-5} mm Hg at 950°C for two hours and was found to have a resistance minimum as shown in Fig. 2. It should be noted that the low resistivity of this sample following the treatment in a reducing atmosphere is an indication of high purity. The results upon reheating this specimen in 10 μ of air at 750°C for two hours are also shown in Fig. 2. The resistivity minimum is no longer observed and the residual resistivity is seen to have decreased approximately by a factor of three.

These results, while not completely understood, indicate the strong influence of trace impurities on the residual resistance and the resistance minimum of copper at low temperatures. It is particularly significant in this regard that the anomalous resistivity minimum is reversible and can be introduced or removed at will by a heat treatment in an appropriate atmosphere. These results would then seem to show clearly that the resistance minimum is not a fundamental property of the copper but a manifestation of the

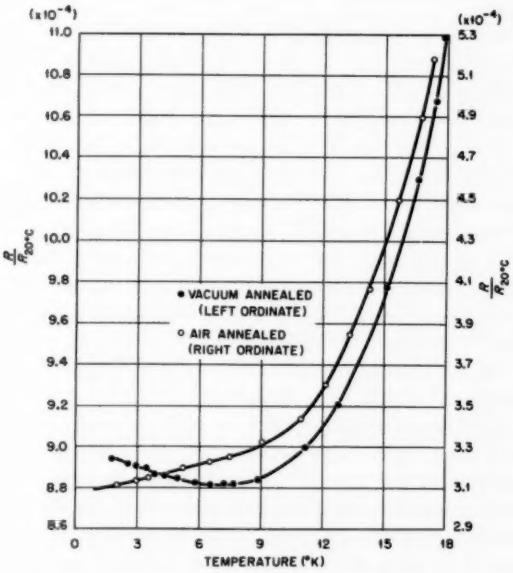

FIG. 2. The temperature dependence of the resistivity of 99.999% pure copper (American Smelting and Refining) after annealing in a reducing atmosphere and an oxidizing atmosphere.

state in which trace impurity atoms are present in the copper. Moreover, these results suggest that the many conflicting observations on the resistance minimum may be due to differences in the preparation of the samples.

In addition to work now in progress with dilute alloys of copper with various impurities subjected to oxidizing and reducing atmospheres, a similar study is contemplated for silver and gold. The effect of other reducing atmospheres on the resistivity of copper is also under investigation.

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[1]D. K. C. MacDonald and W. B. Pearson, Acta Met. 3, 392 (1955).

[2]Blewitt, Coltman, and Redman, Phys. Rev. 93, 891 (1954).

[3]Redman, Coltman, Blewitt, and Klabunde, Bull. Am. Phys. Soc. 4, 150 (1959).

MAGNETIC SPIN PUMPING IN FLUIDS CONTAINED IN POROUS MEDIA*

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In several cases a large nuclear polarization has been produced in a fluid by pumping with microwave radiation in resonance with paramagnetic centers near a solid-fluid interface.[1,2] The number of such paramagnetic sites per unit volume of the sample is often very small. Only nuclei in the fluid molecules nearest to the paramagnetic centers are directly experiencing the spin flips which give rise to such an Overhauser-type effect. Therefore, molecular diffusion must play an essential role in these phenomena. In this note we derive a theoretical expression for the total nuclear polarization, which contains the fluid volume as an independent variable.

Let an amount of fluid with volume V and of spherical shape be bounded by the solid. The surface contains a number of paramagnetic centers which are in resonance with the microwave power; we denote by V_p the volume occupied by fluid molecules in the surface layer in the immediate vicinity of these centers, and by V_s (which is part of V) the volume occupied by fluid molecules in the remainder of the surface layer. We will assume that $V_p \ll V$. The nuclei in V_p are flipped at a given rate by the external microwave power through the two known processes of the Overhauser type or through that of the type reported by Uebersfeld et al.[1] and by Abragam and Proctor.[3] Some flipped nuclei escape to the volume V before they are relaxed. In V they are exposed to the ordinary relaxation, characterized by the relaxation time T_1^0. They also may penetrate the part V_s of V, as we assume that the fluid forms a single continuous phase. In V_s relaxation is influenced by the strongly reduced mobility of the molecules which, unless one approaches a solid-like condition, is an enhancement. Also surface magnetism not in resonance with the microwave power can contribute to the relaxation in V_s. We denote by T_1^s the relaxation time in V_s. Finally the nuclei may move back into V_p and be relaxed there, with a characteristic time T_1^p.

In a steady situation a complicated pattern of nuclear polarization will be established, given by the density $m_0 + m(x, y, z)$, where m_0 is the equilibrium value. We wish to calculate the total excess magnetization, $M = \int_V m dx dy dz$. This can be obtained from two simultaneous equations, for the steady state spin polarization in V and in V_p, respectively. The coupling describes the transfer of spin polarization across their boundary. In V

we use the equation[4] for nuclear relaxation with diffusion:

$$0 = \partial m/\partial t = D\nabla^2 m - m/T_1', \qquad (1)$$

where D is the molecular diffusion constant, and T_1' is the relaxation time in V excluding the contribution from V_p. One of us[5] has shown that under a wide variety of circumstances in sufficiently small pores:

$$1/T_1' = 1/T_1^0 + (V_S/V)/(T_1^S + \tau_S), \qquad (2)$$

where τ_S is the sticking time of molecules entering V_S. As a boundary condition for (1) we use the condition that the flux of polarization across the surface of the sphere equals the net flux across the boundary of V_p and V:

$$4\pi R^2 D(dm/dr)_{r=R} = (V_p/\tau_p)[m_p - m(R)], \qquad (3)$$

where R is the radius of the sphere, m_p is the steady state polarization density in V_p, and τ_p is the sticking time of molecules in V_p. Finally m_p is determined by the production rate of nuclear polarization in V_p, and by local relaxation and net transfer to V:

$$0 = dm_p/dt$$

$$= \rho K S^*/T_1^p - m_p/T_1^p - [m_p - m(R)]/\tau_p. \qquad (4)$$

Here ρ is the nuclear density in V_p, S^* is a measure for the electron spin polarization produced by the microwave source: $S^* = S_z - S_0$, i.e., the deviation of spin polarization from thermal equilibrium, for the Overhauser effect and $S^* = S_z$ for the effect reported in references 1 and 3. $K = 1$ for the effect mentioned last under the conditions of their experiments and for the Overhauser effect due to contact interaction; $K = -1/2$ for the Overhauser effect due to dipole-dipole interaction.

The solution of (1) which is well behaved at $r = 0$ is of the form:

$$m(r) = C \sinh u/u, \quad u = r(DT_1')^{-1/2}.$$

Using (3) and (4), we obtain in the limit that $R(DT_1')^{-1/2} \ll 1$:

$$M = V_p \rho K S^* T_1/(T_1^p + \tau_p), \qquad (5)$$

where

$$1/T_1 = 1/T_1' + (V_p/V)/(T_1^p + \tau_p).$$

This limiting condition which makes the result independent of the shape, is also a requirement for the validity of (2). Equation (5) also holds obviously when V is very large and at the same time $T_1 \approx T_1^0$. T_1 as given by (6) is, by analogy with (2), seen to be the experimentally observable relaxation time of the fluid in the pore.

One can distinguish two cases:

Case 1. $T_1 \ll T_1^0$:

$$M = V\rho K S^* \left[1 + \frac{V_S}{V_p}\left(\frac{T_1^p + \tau_p}{T_1^S + \tau_S}\right)\right]^{-1}, \qquad (7)$$

Case 2. $T_1 \approx T_1^0$:

$$M = V_p \rho K S^* T_1^0/(T_1^p + \tau_p). \qquad (8)$$

Case 1, which applies when the surface relaxation dominates, describes an enhancement of the nuclear resonance absorption by a factor independent of V. One obtains the full Overhauser effect if relaxation in V_p dominates, $V_S/(T_1^S + \tau_S) \ll V_p/(T_1^p + \tau_p)$. If relaxation in V_S dominates, one finds a reduced effect. This may be realized, e.g., if the electron resonance is inhomogeneously broadened. In case 2, i.e., when the bulk relaxation dominates, M is independent of V so that the observed enhancement factor varies as V^{-1}. This may apply when V is made very large compared with V_p and V_S, or when τ_p and τ_S are very long. One may conclude that the study of spin pumping in these systems gives rather detailed information concerning relaxation at the surface.

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*Work was performed in the La Habra Laboratory of the California Research Corporation.

[1]Erb, Motchane, and Uebersfeld, Compt. rend. 246, 2121 (1958).

[2]J. P. Borel and P. Cornaz, Compt. rend. 247, 1988 (1958).

[3]A. Abragam and W. G. Proctor, Compt. rend. 246, 2253 (1958).

[4]H. C. Torrey, Phys. Rev. 104, 563 (1956).

[5]H. C. Torrey (to be published).

NOVEL METHOD OF SPECTROSCOPY WITH APPLICATIONS TO PRECISION FINE STRUCTURE MEASUREMENTS

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A new spectroscopic method which promises to yield very precise measurements of some atomic fine structure intervals is described. This technique exploits interference phenomena that can occur in the resonance fluorescence of an atom in which two of the excited Zeeman substates cross.

The method will be described in reference to the apparatus utilized for the determination of the helium $2\,^3P_1 - 2\,^3P_2$ separation. See Fig. 1(a). A beam of helium light is projected through a helium discharge tube and detected by a PbS cell sensitive to the helium 1-micron line. The discharge tube contains ~0.2 mm of pure He and is situated between the poles of a Varian 12-in. magnet. The discharge is maintained by a high-voltage radio-frequency source and is rather weak for best results.

With this apparatus a distinct increase in the light received by the detector is found at field strengths of 562.8 and 578.6 gauss. These signals can be displayed on the cathode-ray oscilloscope with a signal-to-noise ratio of ~100:1, and a line width of 2.7 gauss. See Fig. 1(b). The effects described can be observed with just the light from the discharge tube itself. However, the signal-to-noise is very poor because of the very low light intensity. Signals near 1300 gauss can also be observed if the incident light is appropriately polarized.

The explanation of these signals lies in the interference phenomena resulting from a crossing of two of the ^3P Zeeman sublevels. See Fig. 2. If the levels are all distinct (i.e., separations greater than their natural widths), then each contributes separately to the resonance scattering. However, when two levels are degenerate, their contributions to the scattering may interfere,

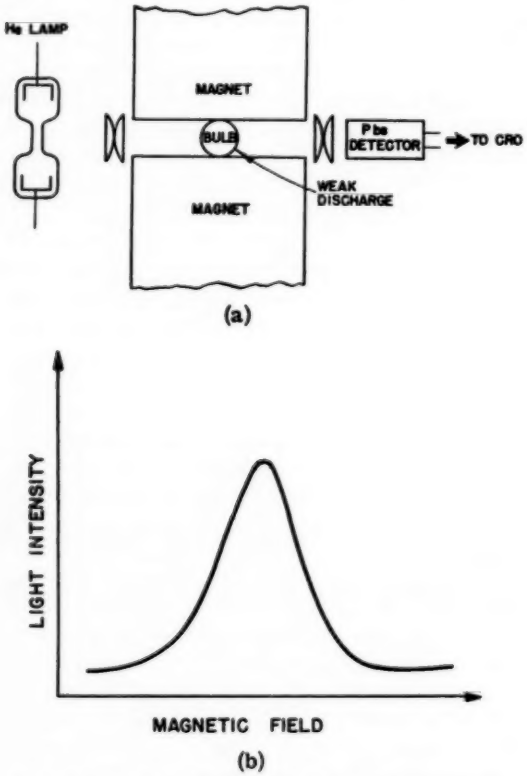

FIG. 1. (a) A schematic diagram of the apparatus employed in the helium experiment. (b) One of the $\Delta m = 2$ (600 gauss) lines as displayed on an oscilloscope.

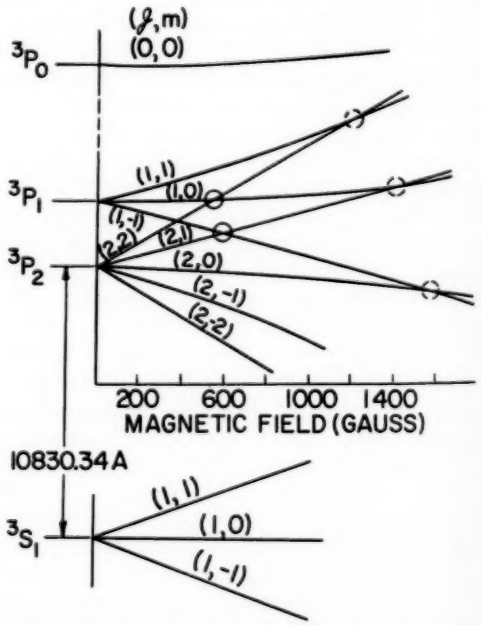

FIG. 2. A sketch of the helium energy levels pertinent to the $n = 2$ fine structure measurement.

thus giving rise to a change in scattered light.

In order to illustrate the calculation of this effect, let us consider a system having one ground state A and two excited states B and C. (A corresponds to one of the $^3S_1$ levels and B and C correspond to two of the ^3P levels.) To calculate the resonance fluorescence of this system, one needs first to calculate the rate at which photons of polarization $\vec{e}_1$ are absorbed and photons of polarization $\vec{e}_2$ are re-emitted. For the case where the levels B and C are well resolved (i.e., separated by more than the natural line width), one has

$$R \sim |(A|\vec{e}_2\cdot\vec{r}|B)(B|\vec{e}_1\cdot\vec{r}|A)|^2 + |(A|\vec{e}_2\cdot\vec{r}|C)(C|\vec{e}_1\cdot\vec{r}|A)|^2. \quad (1)$$

For the case where B and C have the same energy,

$$R \sim |[(A|\vec{e}_2\cdot\vec{r}|B)(B|\vec{e}_1\cdot\vec{r}|A) + (A|\vec{e}_2\cdot\vec{r}|C)(C|\vec{e}_1\cdot\vec{r}|A)]|^2. \quad (2)$$

It is clear that in order for Eqs. (1) and (2) to yield different results both terms in each expression must be nonvanishing; i.e., each of the levels B and C must be able to share the same photon. For the case of unpolarized light, one finds that the two degenerate levels must differ in m by 0 or 2 in order for there to be an interference term. With linear polarized light, one finds that crossovers of levels differing in m by 1 can be observed provided the direction of polarization is not parallel or perpendicular to the magnetic field. Levels differing in m by 3 or more will not interfere. By summing over the directions of $\vec{e}_2$, it can be shown that the total resonance scattering rate is not affected by the crossover of the two levels; only the angular distribution is changed.

This mechanism leads to a qualitative understanding of the observed signals. The two signals near 600 gauss are due to the crossing of levels with $\Delta m = 2$; signals near 1300 gauss are due to the crossing of levels with $\Delta m = 1$. No signal is observed near 400 gauss at the crossing of levels with $\Delta m = 3$. The observed line width is in approximate agreement with the calculated mean lifetime of the ^3P levels, and the intensity of the signal is in approximate agreement with that computed from the geometry of the apparatus and the relevant matrix elements.

The signal observed at 578.6 gauss arises from the crossover of the (2, 1) and (1, - 1) levels. The magnetic field at which this crossover occurs depends only upon the fine structure splitting $^3P_1 - {}^3P_2$ and well-known physical constants. By measuring the magnetic field, then, this splitting can be determined. The signal has been observed in several different bulbs varying in pressure from 0.16 to 1.5 mm and under different discharge conditions. There is no evidence, within our accuracy, of a pressure shift or a sensitivity to discharge conditions. We find for the splitting:

$$^3P_1 - {}^3P_2 = 2291.56 \pm 0.09 \text{ Mc/sec.}$$

The major contribution to the error interval arises from the inhomogeneity of the magnetic field over the discharge tubes which had volumes of several cubic inches. Experiments in progress will utilize smaller volumes. Our result is to be compared with the only other reported precision measurement, that of Wieder and Lamb[1]:

$$^3P_1 - {}^3P_2 = 2291.72 \pm 0.35 \text{ Mc/sec.}$$

The magnetic field at which the (2, 2) - (1, 0) crossover occurs depends slightly on the $^3P_1 - {}^3P_0$ separation. Using our determination of the $^3P_1 - {}^3P_2$ separation and the measured magnetic field of this crossover, we find

$$^3P_0 - {}^3P_1 = 29\,650 \pm 280 \text{ Mc/sec.}$$

The large error indicates the slightness of the field dependence of the crossover on this separation. This determination is to be compared with the only measurement of comparable accuracy, the optical determination of Brochard et al.[2]:

$$^3P_0 - {}^3P_1 = 29\,640 \pm 300 \text{ Mc/sec.}$$

It would be very desirable to measure the crossover of the (0, 0) and (2, 2) levels, which occurs in the neighborhood of 8300 gauss, because this would yield a very accurate measurement of the $^3P_0 - {}^3P_1$ separation. A preliminary search for the signal due to this crossover was made without success because the relatively simple discharge geometry is very susceptible to plasma oscillations at fields in excess of 3000 gauss. Work is in progress on discharge designs that should obviate this difficulty.

The method discussed in this Letter should be applicable to the measurement of the fine structure splitting $^2P_{3/2} - {}^2P_{1/2}$ in hydrogen, as well as other fine and hyperfine structure separations. Experiments along these lines are in progress.

A detailed analysis of the effects so briefly

described in this Letter is in preparation and will be submitted to the Physical Review.

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[1]I. Wieder and W. E. Lamb, Phys. Rev. 107, 125 (1957).

[2]Brochard et al., J. phys. radium 13, 433 (1952).

MAGNETIC DIPOLE RESONANCE OF EXCITED LEVELS IN FREE IONS OF Cd II AND Zn II

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(Received September 28, 1959)

In exciting by electron impact free Cd and Zn atoms in an atomic beam, one also obtains ionic excited levels. In particular, if an electron is pulled out of the d shell of the atomic ground state, $^1S_0\ d^{10}s^2$ ($3d^{10}4s^2$ and $4d^{10}5s^2$ for Zn and Cd, respectively), this leads directly to the ionic excited level, the inverted doublet, $^2D\ d^9s^2$. The only ionic levels of lower energy are the ionic ground state $^2S_{1/2}\ d^{10}s$, and the doublet $^2P\ d^{10}p$.

The observed transitions,[1] from the $^2D\ d^9s^2$ doublet to lower states, are the two-electron jumps $^2D \rightarrow {}^2P$:

	For Cd II	For Zn II
$^2D_{1/2} \rightarrow {}^2P_{3/2}$	4416 A	7479 A
$^2D_{3/2} \rightarrow {}^2P_{3/2}$	3536 A	6215 A
$^2D_{3/2} \rightarrow {}^2P_{1/2}$	3250 A	5894 A

We have observed the magnetic dipole resonance of the 4416A Cd II line and of the 5894A Zn II line by measuring polarization differences $I_\pi - I_\sigma$ as a function of the steady magnetic field. The experimental arrangement and method of observation were similar to the classical experiment of the Kastler-Brossel[2] type, except that in our case the excitation was by electron impact instead of an optical method.

We conclude that the $^2D\ d^9s^2$ doublet Zeeman sublevels become unequally occupied through this ionization process.

We have determined lifetimes of the $^2D_{5/2}$ state of Cd II and the $^2D_{3/2}$ state of Zn II by the method given in Brossel's paper,[2] and the polarization percentage of the 5894A line of Zn II.

The mean lifetimes are:

$$^2D_{5/2} \text{ of Cd II:} \quad T_e = (8.3 \pm 0.7) \times 10^{-7} \text{ sec},$$

$$^2D_{3/2} \text{ of Zn II:} \quad T_e = (4.65 \pm 0.2) \times 10^{-7} \text{ sec}.$$

It is of interest to note that these lifetimes are relatively long compared to lifetimes of 10^{-8} - 10^{-9} sec of levels corresponding to allowed transitions. It is reasonable that a two-electron jump has a longer lifetime than a one-electron jump. Incidentally, we note that a one-electron jump with $\Delta l = 2$ (electric quadrupole transition) is 10^6 times less probable than a $\Delta l = 1$ transition (electric dipole transition), and therefore still 10^3 - 10^4 times less probable than the two-electron jump transition we observed.

Nevertheless, the two observed lines are still as strong as allowed atomic transitions. We assume that the reason for this high intensity is a large d-shell ionization cross section. Besides, to arrive at the $^2D\ d^9s^2$ state, the processes of ionization and excitation necessarily occur in one step.

Figure 1 shows the polarization percentage of the 5894A line of Zn II versus electron energy, compared to the excitation function; the polarization percentage being defined as $p = 100(I_\pi - I_\sigma)/(I_\pi + I_\sigma)$. We deduce for the optimum population ratio of the $^2D_{3/2}$ level Zeeman sublevels (corresponding to 8% polarization):

$$\text{population } (\tfrac{3}{2})/\text{population } (\tfrac{1}{2}) = 0.805.$$

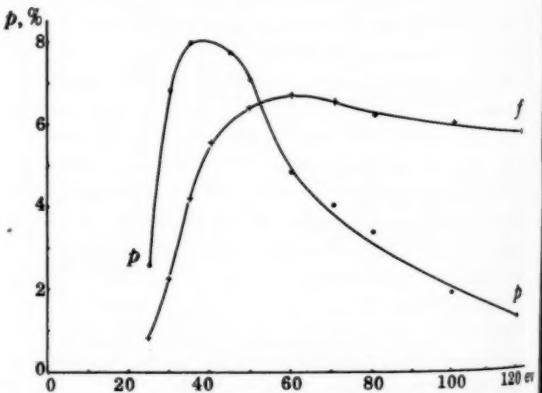

FIG. 1. Percentage polarization p in % and excitation function f in arbitrary units of the 5894A line of Zn II versus electron energy in electron volts.

To calculate the ratio, we have used intensity formulas for Zeeman components in a Russell-Saunders scheme.

We have seen no resonance of the 3250A and 3536A lines of Cd II. Possibly, the polarization of the $^2D_{3/2}$ state of Cd II is too low.

With our detection methods we have not been able to observe the 6215A and 7479A lines of Zn II.

We believe that this is the first time that magnetic dipole resonance of free ions has been observed.

The observation of the magnetic resonance of atomic ions might, possibly, be of use for the determination of local magnetic fields in a plasma containing these ions as impurities.

[1]Y. Takahashi, Ann. Physik 3, 27 (1929); K. Larché, Z. Physik 67, 465 (1931).

[2]A. Kastler, Physica 12, 619 (1946); Physica 17, 191 (1951); Proc. Phys. Soc. (London) A67, 853 (1954). F. Bitter and J. Brossel, Massachusetts Institute of Technology Technical Report No. 176, 1950 (unpublished); J. Brossel, thesis, Paris, 1951 (unpublished); J. Brossel and F. Bitter, Phys. Rev. 86, 308 (1952).

ANOMALOUS LOSS OF RESOLUTION OF PARAMAGNETIC RESONANCE HYPERFINE STRUCTURE IN LIQUIDS*

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(Received October 1, 1959)

Hausser[1] has recently observed a remarkable temperature dependence in the resolution of hyperfine splittings for paramagnetic molecules in liquid solution. Typically, he finds that increasing temperature from a point of high solvent viscosity ultimately brings out a well-resolved structure which surprisingly blurs and disappears at still higher temperatures. We propose here an explanation for the anomalous loss of structure.

Invoking theories of motional narrowing,[2-4] one can understand the initial appearance of structure as temperature increases by recognizing that the correlation time for molecular re-orientation,

$$\tau_C = 4\pi\mu a^3/(3kT), \qquad (1)$$

decreases until its reciprocal is comparable to the frequency magnitude of anisotropic hyperfine splittings, which are then averaged out by the molecular tumbling.[5] The characteristic isotropic hyperfine pattern then emerges. On this supposition, Eq. (1), together with the solvent viscosity μ appropriate to the lowest temperature at which structure appears and $(2\pi\tau_C)^{-1}$ comparable to the expected anisotropic coupling, yields reasonable values of a, the equivalent Stokes' law spherical radius for the free-radical molecule in solution.

The motional narrowing theory, which predicts for decreasing τ_C a monotonic decrease of each hyperfine component's width (including static and dynamic contributions[4]), cannot explain the loss of hyperfine resolution with further temperature increase. We suggest instead that this phenomenon arises from smearing out of the isotropic hyperfine structure by the exchange interaction, as in the concentration dependence of hyperfine resolution in solutions of diphenyl picryl hydrazyl (DPPH).[6]

We suppose the following model. A paramagnetic solute molecule in moderately low concentration swims through the solvent ultimately into the vicinity of a like solute molecule. If the two approach closely enough for appreciable overlap of their unpaired electron distributions, the exchange interaction $-2J\vec{S}_1\cdot\vec{S}_2$—is "turned on." Assuming that the exchange integral J exceeds the Zeeman interaction $g\beta H_0$ with the external magnetic field, the electron spins will undergo appreciable relative precession about each other, provided they hover long enough in close interaction. The phases of their Larmor precessions about H_0 will have been interrupted, and when the interruption rate is comparable to the width of the hyperfine components, they will be broad-

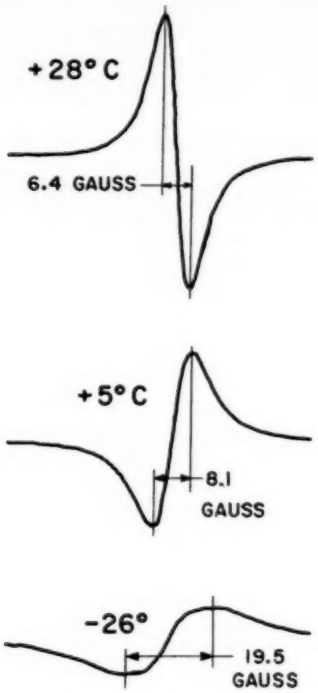

FIG. 1. Temperature dependence of exchange narrowing in 0.14*M* DPPH-toluene solution. The resonance is observed at 9.2 kMc/sec.

ened and the structure will begin to blur.

Figures 1 and 2 show spectra observed at 9.2 kMc/sec in a 0.14*M* solution of DPPH in toluene. It is clear that further temperature increases above -41°C, where the structure has already begun to smear out, lead to further narrowing even of the single resonance, as observed with exchange narrowing when concentration increases.[6,7]

Our analysis of this model is crude but perhaps instructive. If we suppose the diffusion of the free-radical solute molecule to take place in steps of rms length λ from one molecular environment to another at an average rate of n steps per second, the diffusion constant D is [8]

$$D = n\lambda^2/6. \tag{2}$$

If the average number of new molecular neighbors encountered with each step is z, the number of molecules encountered per unit time is zn. A fraction N_γ/N_S of these are other solute molecules, where N_γ and N_S are the volume concentrations of free-radical and solvent molecules. Therefore, the rate at which a paramagnetic

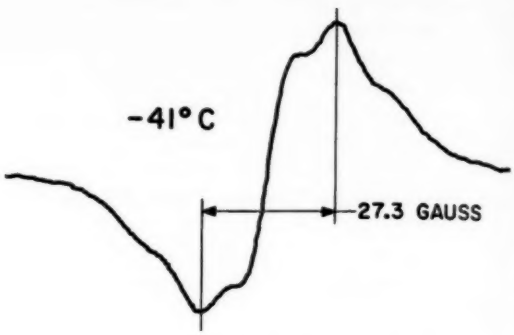

FIG. 2. Emergence of hyperfine structure in the spectrum of a 0.14*M* DPPH-toluene solution at -41°C.

molecule encounters another one is znN_γ/N_S. If p is the probability that appreciable spin precessions about each other occur upon such encounters, the frequency of phase-interrupting collisions is

$$f_e = znN_\gamma p/N_S = 6zN_\gamma Dp/(\lambda^2 N_S). \tag{3}$$

Because N_γ will be fairly small, N_S^{-1} may be replaced by the solute molecular volume $(4\pi/3)a_S^3$, a_S being the equivalent spherical radius of a solute molecule. D is related to the viscosity μ through the Stokes-Einstein relation,

$$D = kT/(6\pi a_\gamma \mu). \tag{4}$$

Then,

$$f_e = \frac{N_\gamma kT}{\mu}\left[\frac{4zpa_S^3}{3\lambda^2 a_\gamma}\right]. \tag{5}$$

The dimensionless bracket involves ratios of quantities comparable to molecular radii. The probability p depends upon the time t_i of interaction between paramagnetic molecules, which must be comparable to the period $\hbar/J$ of mutual spin precession about each other if appreciable mutual precession and thus phase interruption of the Larmor precession are to occur. Presumably, $t_i \approx 1/n$. For $\mu = 10^{-2}$ poise, Eqs. (2) and (4) with reasonable values of λ and a_γ yield $1/n$ in the range of 10^{-10} to 10^{-11} sec, which is comparable to $\hbar/J$ during the interaction. (J is estimated for close interaction distance from exchange narrowing in crystalline DPPH.) We can only guess that p is not very small and that the bracket in Eq. (5) should be near unity.

For DPPH the splitting to be obscured is about $\Delta\nu = 30$ Mc/sec, and a component width half that, corresponding to $f_e \approx 1.5\times10^7$, would effectively remove structure. From Figs. 1 and 2, this

circumstance nearly obtains at -41°C in 0.14*M* solution. Since $\mu = 1.5\times10^{-2}$ poise, the bracket becomes

$$[\ ] = \mu f_e/(N_\gamma kT) \approx 10^{-1}.$$

The same solution's spectrum at +28°C should again yield the bracket. The exchange-narrowing theory[4] relates the observed width $\delta\nu$ of the single narrowed line to the second moment $\langle\Delta\nu^2\rangle$ of the hyperfine pattern and f_e as

$$\delta\nu \approx \langle\Delta\nu^2\rangle/f_e\ . \qquad (6)$$

The value of $\delta\nu$ corresponding to 6.4 gauss and $\langle\Delta\nu^2\rangle$ corresponding to the measured[9] 180 gauss[2] thus fix f_e as about 8×10^7 sec^{-1}. The viscosity of toluene at 28°C and Eq. (5) then yield about 10^{-1} again for the bracket.

Hausser's data on $10^{-3}M$ α,γ-bisdiphenylene β-phenyl allyl in ethanol at 0°C give the bracket a value 2 for that system. Considering the crudeness of our model, including its spherical approximation for aromatic molecules, the bracket values lend credence to the suggested mechanism. If less complicated systems can be found with which to test these ideas, they might merit theoretical refinement.

We wish to thank Mr. John Goldsborough and Professor L. I. Schiff for helpful discussions.

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[1]K. H. Hausser, Z. Naturforsch. 14a, 425 (1959).
[2]Bloembergen, Purcell, and Pound, Phys. Rev. 73, 679 (1948).
[3]P. W. Anderson, J. Phys. Soc. Japan 9, 316 (1954).
[4]R. Kubo and K. Tomita, J. Phys. Soc. Japan 9, 888 (1954).
[5]S. I. Weissman, J. Chem. Phys. 22, 1378 (1954).
[6]Hutchison, Pastor, and Kowalsky, J. Chem. Phys. 20, 534 (1952).
[7]See Fig. 2 on p. 329 of reference 3. Although the model Anderson calculates is a simplified one, it provides an illustration of the broadening of each fine structure component preliminary to a narrowing of the entire pattern into a single line. As the mean interruption frequency further increases, the single line itself grows narrower.
[8]S. Chandrasekhar, Revs. Modern Phys. 15, 1 (1943).
[9]J. Herve, Arch. science (Geneva) 11, 185 (1958).

DISSOCIATION OF H_2^+ IONS BY HYDROGEN

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Interest in the dissociation of H_2^+ ions by hydrogen and other gases has been stimulated recently by the injection problems of thermonuclear devices and accelerators. In particular, the injection current required for a thermonuclear experiment such as OGRA is critically dependent on the cross section for this process, yet there is a discrepancy of a factor 3 between the values obtained by different workers. In some of this work, the importance of the charge exchange cross section and the cross section for double proton production at energies of about 100 kev does not appear to have been realized and none of the techniques so far used has enabled all these processes to be resolved.

In the present work, the cross sections for the four processes contributing significantly to loss of the H_2^+ ions are separately determined. These processes are:

$$H_2^+ - H^0 + H^+, \qquad (1)$$

$$H_2^+ - H^+ + H^+ + e, \qquad (2)$$

$$H_2^+ - H^0 + H^0 - e, \qquad (3)$$

$$H_2^+ - H_2^0 - e, \qquad (4)$$

with cross sections σ_1 to σ_4, respectively.

H_2^+ ions accelerated in a 3-Mv Van de Graaff machine were passed through a differentially pumped gas chamber. The beams of neutral particles, H_2^+ ions, and protons were then separated by a magnetic field and detected by 1-in. CsI crystals mounted on photomultiplier tubes. The entry of two protons simultaneously into the crystal from process (2) was recorded as a pulse of double the height of that from a single proton [process (1)]. Similarly processes (3) and (4) produced pulses in the neutral particle counter of double the height of those due to neutral particles from process (1). By differential discrimination of the output from the crystals it was possible to separate very clearly double from single events and hence the partial cross sections for the processes (1), (2), and (3)+(4).

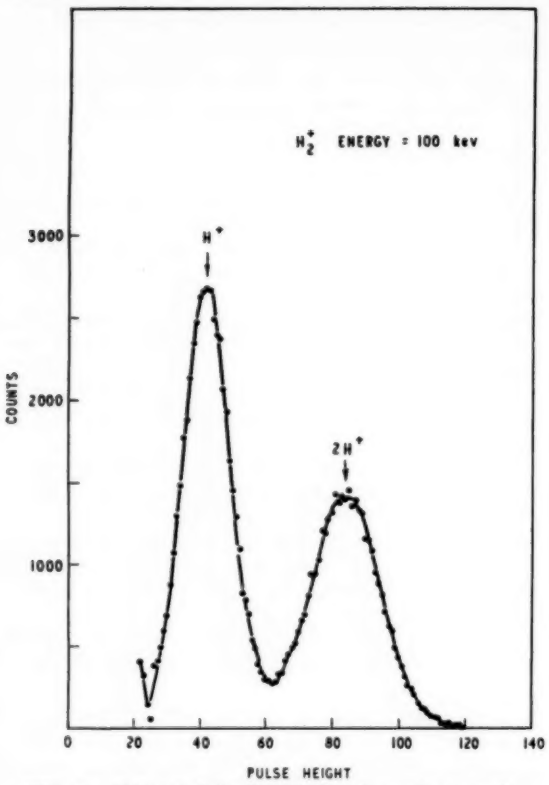

FIG. 1. Pulse-height distribution in proton counter for 100-kev H_2^+ energy. The upper peak represents the two protons from process (2) entering the crystal simultaneously.

Figure 1 shows the pulse-height distribution from the proton counter at 100-kev H_2^+ energy. At higher energies the groups were clearly separated and the peak to valley ratio was about 200:1.

In order to separate (3) from (4) a separate experiment was performed with a narrow slit (0.148 mm across) mounted in front of the neutral-particle detector. The width of the slit was such that the probability of both of the H^0 particles from process (3) entering the crystal was negligible. Under these conditions, the double-height pulses corresponded only to H_2^0 particles from process (4). The slit was moved across the beam and the count rate plotted against position for a given count on the H^+ counter. Integration of the resulting curves gave the cross section for H_2^0 production. In the energy range 100 - 200 kev it was found to be equal to about 30% of $\sigma_3 + \sigma_4$. Good agreement was obtained between total neutral particle counts with and without the slit.

The partial cross section results are shown in Fig. 2. It is seen that the partial cross section

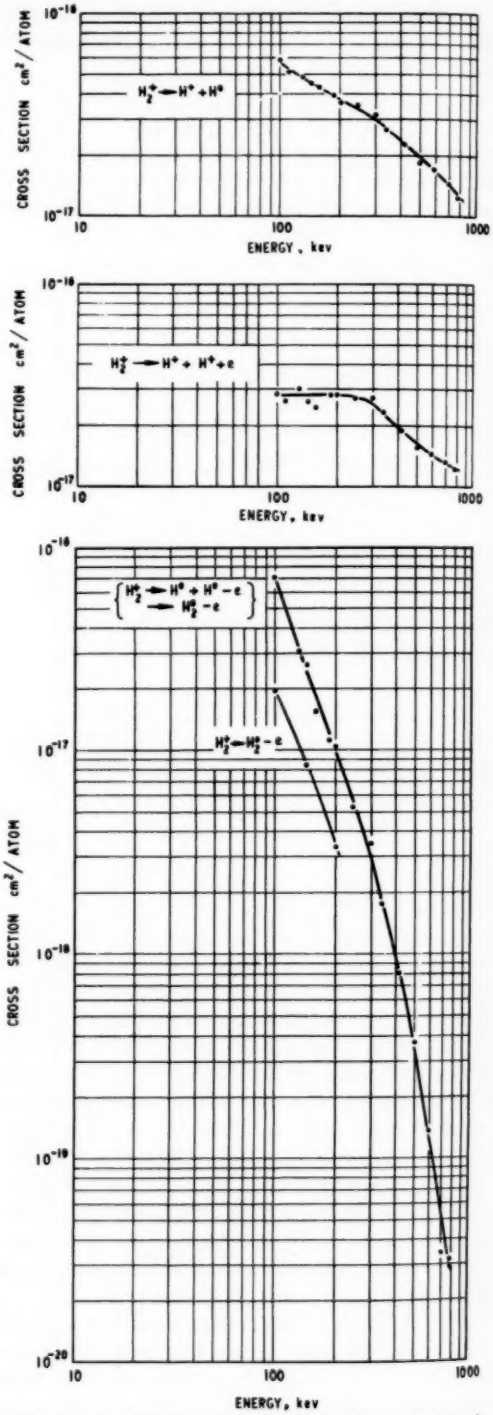

FIG. 2. Partial cross sections for dissociation of H_2^+ into $H^0 + H^+$, $2H^+$, $2H^0$, and H_2^0 plotted against H_2^+ energy. The fraction of the total double neutral particle production remaining as H_2^0 is indicated.

for double proton production is important at all energies and is a slowly varying function as would be expected for excitation to the repulsive double proton state. The partial cross section for double neutral particle production is important at low energies and is a strong function of energy, consistent with a charge exchange mechanism. The angular distribution of the H^0 particles from process (3) is narrower than that from process (1) indicating that process (3) proceeds by charge exchange followed by breakup of 70% of the H_2^0 particles by excitation to the $1\,^3\Sigma_u$ repulsive state.

The total cross section for loss of H_2^+ ions is plotted in Fig. 3 together with the results of other workers. The results of Damodaran[1] may be compared directly with ours and are about 30% higher, as are also those of Barnett[2] at high energies. The low-energy cross sections of Barnett were defined on the assumption that the cross sections for processes other than simple dissociation were small. In view of the large value of these at low energies it is not easy to compare the results directly. However, if in Barnett's work only a small fraction of the particles were dissociated then the cross sections should approximate to those for proton production. These are plotted in Fig. 3 where it is seen that his results are a factor of 1.8 lower than ours. Comparison with the results of Fedorenko[3,4] is not possible until the details of the experimental measurements are known but their cross sections would appear to be somewhat larger than ours.

The principal source of error in the measurements reported here is in reading the pressure in the gas chamber. It is estimated that the results are accurate to ±10% in absolute value.

The author is extremely indebted to Mr. R. E. Bradford for his work in designing the apparatus and taking the results, and to Dr. K. W. Allen for stimulating discussions.

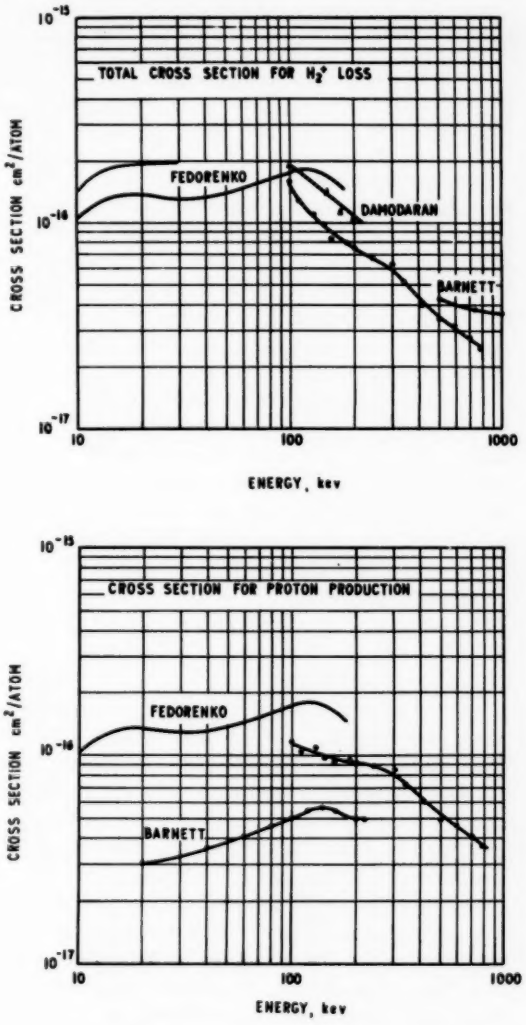

FIG. 3. Total cross section for H_2^+ loss and cross sections for proton production ($\sigma_1+\sigma_2$) plotted against H_2^+ energy.

[1]K. K. Damodaran, Proc. Roy. Soc. (London) A293, 382 (1957).

[2]C. F. Barnett, Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1959), Paper No. P/1789.

[3]N. V. Fedorenko, J. Tech. Phys. 24, 769 (1954).

[4]N. V. Fedorenko, reported by I. V. Kurchatov, Second Geneva Conference, reference 2.

HYPERFINE STRUCTURE OF THE METASTABLE TRIPLET STATE OF HELIUM THREE*

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(Received October 9, 1959)

The metastable triplet state ($1s\,2s$, $^3S_1$) of the He^3 atom is split by a magnetic field into six energy levels which can be designated by the quantum numbers (F, M). The energy difference between the levels $(3/2, -1/2)$ and $(1/2, -1/2)$ has a minimum value at a magnetic field of about 802.5 gauss. According to the Breit-Rabi formula the corresponding frequency, $\nu_{\min}$, is related to the zero-field hyperfine splitting, $\Delta\nu$, by $\Delta\nu = [3/(2\sqrt{2})]\nu_{\min}$. The frequency $\nu_{\min}$ has been determined by the atomic beam magnetic resonance method. Resonance lines having their natural (transit time) widths can be obtained because of the small (quadratic) dependence of the line frequency on magnetic field, and hence a precise determination of $\nu_{\min}$ is possible. Two different kinds of microwave cavity were used in producing the transitions; one was a rectangular cavity operating in the TE_{101} mode with a length of 3.8 cm along the direction (y axis) of beam propagation, and the other was a cylindrical coaxial cavity 2.4 cm long. Resonance lines obtained with these two cavities were, respectively, about 40 and 55 kc/sec wide. Among the parameters varied between determinations were the intensity of the static magnetic field, the intensity of the microwave field, the tuning of the cavities, and the orientation of the cavities with respect to the direction of beam propagation. The final value is $\Delta\nu = (6739.7013 \pm 0.0004)$ Mc/sec. This value overlaps a preliminary value reported on this research[1]; it agrees with an earlier determination[2] of $\Delta\nu$ made with $\Delta M = \pm 1$ transitions at low static field, and is more precise by a factor of 125.

A theoretical expression for the hyperfine splitting in the He^3 atom has been given by Sessler and Foley.[3] About 2×10^{-4} of the splitting arises from nuclear structure effects for which only approximate theoretical estimates are available. Detailed comparison between theory and experiment is possible for the ratio between the hyperfine splitting in the He^3 atom and the hyperfine splitting in the $2\,^2S_{1/2}$ state of the He^{3+} ion, since nuclear structure contributions substantially cancel. Use of our value for $\Delta\nu(He^3, {}^3S_1)$ and the value for $\Delta\nu(He^{3+}, {}^2S_{1/2})$ measured by Novick and Commins[4] gives

$$\left.\frac{\Delta\nu(He^3, {}^3S_1)}{\Delta\nu(He^{3+}, {}^2S_{1/2})}\right|_{\text{expt.}} = 6.2211384 \pm 0.0000012. \quad (1)$$

A theoretical expression for this ratio which is claimed[3] to be accurate to about 1 part in 10^6 is

$$\frac{\Delta\nu(He^3, {}^3S_1)}{\Delta\nu(He^{3+}, {}^2S_{1/2})} = 6(1+\epsilon)\{1 + [\Delta_{\text{Rel}}(\text{atom}) - \Delta_{\text{Rel}}(\text{ion})] + \Delta_{\text{Diamag}}(\text{atom}) + [\Delta_{ED}(\text{atom}) - \Delta_{ED}(\text{ion})] + [\Delta_{\alpha m/M}(\text{atom}) - \Delta_{\alpha m/M}(\text{ion})] + [\Delta_{\text{Nuc}}(\text{atom}) - \Delta_{\text{Nuc}}(\text{ion})]\}. \quad (2)$$

Here $(1+\epsilon)$ is a correction[5] to the Fermi formula for a single electron to take account, nonrelativistically, of the second electron in the helium atom; $[\Delta_{\text{Rel}}(\text{atom}) - \Delta_{\text{Rel}}(\text{ion})] = (5.91 - 8.50)\alpha^2$ is the difference between relativistic corrections[3,6] for the atom and ion, and $\Delta_{\text{Diamag}}(\text{atom}) = 4.3\times10^{-6}$ is a diamagnetic correction.[3] The remaining quantities in parentheses are, respectively, the difference of quantum electrodynamic corrections[7] (of relative order $Z^2\alpha^3$), of radiative corrections[8] of order $\alpha m/M$ (obtained when the nuclear motion is treated covariantly; m = electron mass, M = mass of the He^3 nucleus), and of nuclear structure corrections. These differences presumably do not contribute much more than 1 part in 10^6 to the ratio.

Pekeris has calculated from the Schrödinger equation for helium[9]: $1+\epsilon = 1.0370050$ with an estimated uncertainty of 6 parts in 10^7. Using the value for $1+\epsilon$ in Eq. (2), the value 6.221199 is obtained for the ratio of the hyperfine splittings. The calculated ratio differs from the experimental number by 1 part in 10^5. This difference is considerably greater than the combined uncertainties of the experimental ratio and of the computed value of $1+\epsilon$. At present the most likely source of the discrepancy appears to be inaccuracy in the computation of the relativistic correction, $\Delta_{\text{Rel}}(\text{atom})$, which has an estimated uncertainty of 3 parts per million.

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Foundation, and the Office of Naval Research.

†National Science Foundation Predoctoral Fellow, 1957-58.

[1]White, Drake, and Hughes, Bull. Am. Phys. Soc. 4, 10 (1959).

[2]G. Weinreich and V. W. Hughes, Phys. Rev. 95, 1451 (1954).

[3]A. M. Sessler and H. M. Foley, Phys. Rev. 98, 6 (1955).

[4]R. Novick and E. D. Commins, Phys. Rev. 111, 822 (1958).

[5]The quantity $1+\epsilon = D(0)/32$ is defined by the equation

$$1+\epsilon = [\int |\psi(\vec{r}_1, \vec{r}_2)|^2_{r_2=0} d\vec{r}_1 + \int |\psi(\vec{r}_1, \vec{r}_2)|^2_{r_1=0} d\vec{r}_2] / |\psi(0)|^2,$$

where $\psi(\vec{r}_1, \vec{r}_2)$ is the nonrelativistic $1s\,2s$, $^3S_1$ wave function and $\psi(\vec{r})$ is the nonrelativistic hydrogenic $1s$ wave function for He^+.

[6]G. Breit, Phys. Rev. 35, 1447 (1930).

[7]N. M. Kroll and F. Pollock, Phys. Rev. 86, 876 (1952); Heberle, Reich, and Kusch, Phys. Rev. 101, 612 (1956).

[8]H. A. Bethe and E. E. Salpeter, Handbuch der Physik, edited by S. Flügge (Springer-Verlag, Berlin, 1956), Vol. 35.

[9]C. L. Pekeris (to be published). We are indebted to Professor Pekeris for informing us of this result prior to publication.

MEASUREMENT OF THE NUCLEAR g FACTOR OF Li^8†*

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(Received October 12, 1959)

In many cases the capture of polarized thermal neutrons should result in product nuclei which are appreciably polarized.[1-3] If the product decays by beta emission, the well known asymmetry of the decay electrons from polarized nuclei should be observable unless relaxation processes destroy the polarization before the decay occurs. An asymmetry produced in this way was observed at this laboratory in 1957[1] with Li^8, a nuclide of mean life 1.2 sec.

Observation of the asymmetry in beta emission offers a means of measuring the polarization of very small samples of short-lived nuclei. If established resonance techniques are used to perturb the nuclear polarization due to the capture of polarized neutrons, the precise measurement of nuclear g factors should be possible in several cases which cannot be studied by conventional methods. This note describes the successful application of this idea to the case of Li^8.

The resonance effect used in this work was the destruction of the polarization by an rf field, well known in nuclear magnetic resonance work as "rf saturation."[4] The sample nuclei are created with their polarization along a static magnetic field H_0. An rf field $2H_1$ is impressed at right angles to H_0. As the radio-frequency is made to approach f_L, the nuclear Larmor frequency, the polarization along H_0 is destroyed and with it the beta-emission asymmetry. The measurement of f_L in the known field H_0 yields the g factor directly.

The geometry of the experiment is shown schematically in Fig. 1. The target was a single crystal of LiF. In order to avoid self-shielding due to the large Li^6 cross section, the crystal was

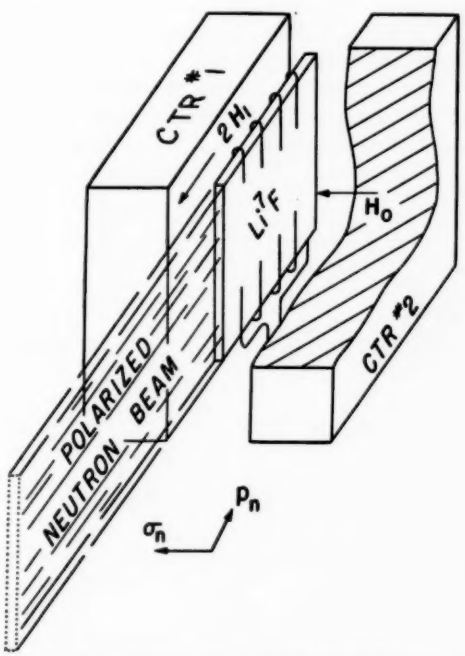

FIG. 1. Schematic experimental arrangement. Beta counters CTR No. 1 and CTR No. 2 detect the electrons emitted with a momentum component parallel or antiparallel to the nuclear polarization. The coil shown around the LiF target provides the rf field $2H_1$ which, at resonance, depolarizes the Li^8 nuclei.

grown from material in which the Li^6 had been depleted to 0.1% of the total Li.[5] The target was irradiated with slow neutrons which had been polarized by reflection from a magnetized cobalt alloy mirror.[6] The neutron polarization, of estimated magnitude 0.8 ± 0.1, was parallel to H_0. Decay electrons with a momentum component along or against H_0 were detected by plastic scintillation counter 1 or 2, respectively. The ratio of counting rates, R_2/R_1, changed by 10% when the neutrons were depolarized by passage through a 0.008-inch steel foil. If the Li^8 ground state has $J^\pi = 2^+$,[7] the observed sign and magnitude of the asymmetry effect require the conclusion that capture occurs almost entirely ($\geq 80\%$) by the $j = 2$ channel and that relaxation of the polarization is negligible.

The rf field, $2H_1$, was provided by the coil wound around the target crystal. When the two counting rates were monitored while the frequency was slowly varied, a narrow range was found in which appreciable depolarization occurred. Subsequent careful observations gave the resonance curve data of Fig. 2. It is interesting to note that only about 20 000 Li^8 atoms existed in the sample at any instant during these measurements. The curve for $2H_1 = 0.08$ oersted shows a width

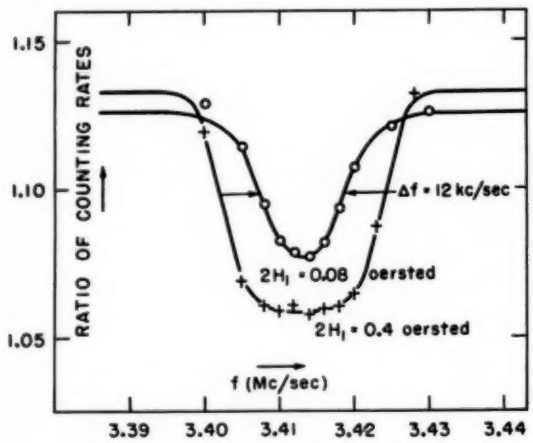

FIG. 2. Nuclear resonance of Li^8 as shown by change of beta-emission asymmetry when the frequency of the rf field is varied. Static field $H_0 = 5418$ oersteds, rf field strength $2H_1$ as shown on curves. The asymmetry effect appears to be 7% rather than 10% as given in the text because no correction has been made for an unusually high background due to pile gammas at the time these data were taken. Other data show definitely that complete depolarization is obtained at resonance with $2H_1 \geq 0.4$ oersted.

of about 0.35% or 19 oersteds in the 5400-oersted static field. In the main, this is accounted for by the local variation in static field due to the magnetic moments of nearby Li^7 and F^{19} nuclei, a width of 13 oersteds for the Li^7 and F^{19} resonances in LiF being found in nuclear magnetic resonance studies.[8] Part of the width may be due to the saturation broadening which clearly appears in the $2H_1 = 0.4$ oersted curve. This broadening may be understood qualitatively when it is realized that the local field at a point fluctuates rapidly (order of 10^4/sec) because of spin flips of adjacent nuclei so that the resonant H value exists for a small fraction of the time even when the external field H_0 is 10 oersteds or more from resonance. Since the time for depolarization is of the order $H_0/f_L H_1$, for sufficiently large H_1 the time at resonance is enough for appreciable depolarization even when the frequency is considerably off resonance in H_0. The inhomogeneity of H_0 was no more than 0.05% over the sample volume so that broadening from this source was negligible.

From the data of Fig. 2, one obtains for the resonance frequency in 5418 ± 1 oersteds (measured with nuclear magnetic resonance techniques), $f_L = 3.413 \pm 0.001$ Mc/sec. The g factor is therefore $g(Li^8) = 0.8265 \pm 0.0004$ nm/h when no diamagnetic corrections are applied. With spin 2,[7] the magnetic moment is $\mu(Li^8) = 1.653 \pm 0.0008$ nm. This g value is very close to that of Li^6 (0.8292). The calculated value for either nucleus in extreme j-j coupling, assuming both the odd neutron and the odd proton to be in $p_{3/2}$ states, is 0.63 or 0.69, corresponding to the use of free-nucleon or "empirical" g factors for the odd nucleons.[9] In LS coupling, for Li^8 the value 0.49 is obtained.[10] (For Li^6, the LS value is 0.85.) With intermediate coupling, Kurath[11] is able to fit the observed value.

Attempts to apply this method to other nuclides are in progress.

I am indebted to T. B. Novey of this laboratory and to V. L. Telegdi of the University of Chicago for having suggested this experiment and for many stimulating discussions.

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†Work performed under the auspices of the U. S. Atomic Energy Commission.

*Based on a thesis submitted to the Department of Physics, the University of Chicago, in partial fulfillment of the requirements for the Ph.D. degree.

[1]Burgy, Davidon, Novey, Perlow, and Ringo, Bull. Am. Phys. Soc. 2, 206 (1957).

[2]D. Kurath, Bull. Am. Phys. Soc. 2, 206 (1957).

[3]General formulas for the polarization and asymmetry are given by F. L. Shapiro, Uspekhi Fiz. Nauk 65, 133 (1958) (unpublished translation by Lydia Venters, Argonne National Laboratory, Lemont, Illinois).

[4]E. R. Andrew, Nuclear Magnetic Resonance (Cambridge University Press, Cambridge, 1955), p. 18.

[5]Lent by Isotopes Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

[6]D. J. Hughes and M. T. Burgy, Phys. Rev. 81, 498 (1951).

[7]F. Ajzenberg-Selove and T. Lauritsen, Nuclear Phys. 11, 1 (1959).

[8]F. J. Low and C. F. Squire, J. Phys. Chem. Solids 5, 85 (1958). These authors give 11 oersteds for the width measured between points for which the second derivative is zero. The corresponding width at half height for a Gaussian shape is 13 oersteds.

[9]R. J. Blin-Stoyle, Theories of Nuclear Moments (Oxford University Press, Oxford, 1957), Chap. 9.

[10]M. E. Rose and H. A. Bethe, Phys. Rev. 51, 205 (1937).

[11]D. Kurath, following Letter [Phys. Rev. Letters 3, 431 (1959)].

MAGNETIC MOMENT CALCULATION FOR Li^8†

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(Received October 12, 1959)

In view of the recent measurement[1] of the nuclear g factor of Li^8, it is of interest to see whether the intermediate-coupling model[2] is consistent with the measurement. The ground state is assumed to be the ($J=2$, $T=1$) state, which is consistent with the experimental evidence and is also the theoretically predicted assignment for the ground state. The calculation has been carried out as a function of the spin-orbit coupling parameter, a/K, for the relative range of nuclear forces given by $L/K=6.8$. These quantities are defined in reference 2.

The resulting values for the magnetic moment are given in Fig. 1, and an intersection[3] of the theoretical curve with the experimental value occurs for $a/K \approx 2.1$. This is consistent with the other evidence[4] for $A=8$, the $M1$ transition width for the 17.6-Mev gamma decay of the ($J=1$, $T=1$) state in Be^8, which leads to a value of $a/K \sim 2.5$. Therefore the intermediate-coupling model is in agreement with the experimental evidence.

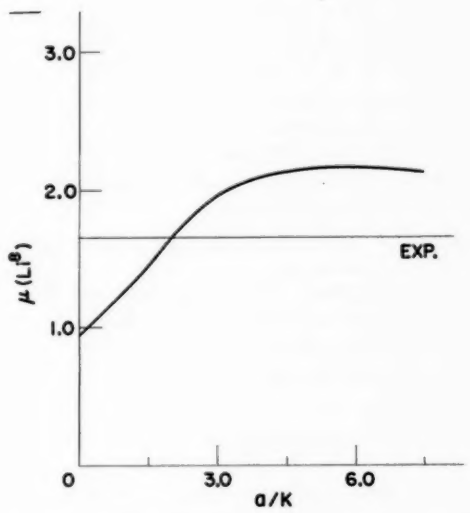

FIG. 1. Magnetic dipole moment of Li^8 in nuclear magnetons as a function of a/K.

†Work performed under the auspices of the U. S. Atomic Energy Commission.

[1]D. Connor, preceding Letter [Phys. Rev. Letters 3, 429 (1959)].

[2]D. Kurath, Phys. Rev. 101, 216 (1956).

[3]There must be at least one more intersection since the value at the jj limit is $\mu=1.25$ nm, but such large values of a/K are not reasonable for a mass number of 8.

[4]D. Kurath, Phys. Rev. 106, 975 (1957).

HIGH NUCLEAR EXCITATION AS A NUCLEAR FORCE PROBE

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(Received September 21, 1959)

In the years since Bethe[1] first derived a statistically based expression for the energy level densities in excited nuclei, particle evaporation experiments at energies in excess of 20 Mev such as that of Eisberg and Igo[2] have shown a disappointing fit to theory. Temperatures deduced from particle evaporation spectra are too low, or what amounts to the same thing, the energy level densities apparently increase faster with excitation energy then the simple theory based on a square or harmonic oscillator well predicts. It is the purpose of this short note to point out (1) that this is to be expected from actual nuclei because of the diffuseness of the actual nuclear potential well and (2) that level densities of highly excited nuclei are a sensitive measure of the details of the shape of the nuclear potential well.

As an example of the effect of the shape of the nuclear potential well on nuclear temperature and energy level density, a calculation was carried out using the diffuse well shape adopted by Ross, Mark, and Lawson[3] to fit Barschall's[4] low-energy neutron scattering data:

$$V(r) = V_0/\{1+\exp[b(r-a)]\}. \quad (1)$$

Using the WKB approximation and adopting the empirically fitted constants of Ross et al., the individual-particle level density above the Fermi level was found to be conveniently approximated by the formula

$$\varphi(\epsilon) = c\epsilon^{1/2}[1+0.02(\epsilon-\epsilon_f)^2], \quad (2)$$

where ϵ is expressed in Mev and the first term alone is the expression for the square well.

Closely following the development of the electron theory of metals[5] and assuming a free nucleon gas in the potential well given by Eq. (1), the excitation energy as a function of gas temperature was easily derived:

$$Q \approx \left(\frac{\pi}{2}\right)^2 \frac{N}{\epsilon_f}(kT)^2 - \frac{3}{5}\left(\frac{\pi}{2}\right)^4 \frac{N}{\epsilon_f^3}(kT)^4 + 0.171\frac{N}{\epsilon_f}(kT)^4$$

$$\equiv \alpha(kT)^2 + \beta(kT)^4, \quad (3)$$

where kT and all other energies are expressed in Mev, N is the number of nucleons, and ϵ_f is the Fermi energy, the energy of the last filled state at zero temperature. The first two terms result from a square well. The third term resulting from the nuclear lip is one hundred times as large as the second term which was justifiably ignored in previous work. Contrary to the square well $(kT)^4$ dependence, the nuclear lip introduces a sizable fourth power dependence on the temperature as illustrated in Table I for $N=100$. Bethe[1] had treated the dependence of nuclear level density on temperature in a more general way than for the quadratic dependence customarily adopted, by calculating the nuclear level density for an arbitrary power relationship between Q and T. Similarly Rosenzweig[6] has recently estimated the nuclear level density for an arbitrary power relationship between $\varphi(\epsilon)$ and ϵ. The overall nuclear level density is found by taking the inverse Laplace transform of the Helmholz free energy[7] which in turn was easily derived from Eq. (3):

$$\rho(Q) = \alpha^{1/4}\pi^{1/2}Q^{-3/4}\exp[2(\alpha Q)^{1/2} + \tfrac{1}{3}\beta(Q/\alpha)^{3/2}]. \quad (4)$$

For β and α derived from 1-3 Mev neutron scattering experiments, the additional term in the exponential would yield a factor of 35 in the level density at excitation energies of 50 Mev, a factor of only 2.4 at 20 Mev.

Several details such as separate wells for neutrons and protons, orbital momentum effects, pairing, and spin-orbit coupling should be incorporated into this simple theory; but the primary effect of the nuclear well shape appears straightforward. Precise determinations of particle spectra should prove very useful in determining the shape and extent of the over-all nuclear force field. The usefulness of the data increases rapidly with excitation energy and indeed, the method is limited to excitation energies in excess of 20 Mev.

It is hoped that this correction to the high-

Table I. The rise in calculated temperature with excitation energy for a diffuse well as compared with that for a square potential well.

Q (Mev)	7	14	28	44	63
kT (square well)	1.0	1.4	2.0	2.5	3.0
kT (diffuse well)	0.94	1.3	1.7	2.0	2.3

energy theory will extend the excellent recent interpretation of thermal neutron cross sections by Rosenzweig,[6] Bloch,[8] and Ross.[9] Particle spectra from highly excited nuclei are currently being interpreted with a more elaborate theoretical treatment in order to arrive at the shape of the nuclear potential.

[1]H. A. Bethe, Revs. Modern Phys. 9, 69 (1937).
[2]R. M. Eisberg and G. J. Igo, Phys. Rev. 93, 1039 (1954).
[3]Ross, Mark, and Lawson, Phys. Rev. 102, 1613 (1956); 104, 401 (1956).
[4]H. H. Barschall, Phys. Rev. 86, 431 (1952).
[5]R. H. Fowler and E. A. Guggenheim, Statistical Thermodynamics (Cambridge University Press, Cambridge, 1952), Chap. XI, pp. 452-458.
[6]N. Rosenzweig, Phys. Rev. 105, 950 (1957); 108, 817 (1957).
[7]I. N. Snedden and B. F. Touschek, Proc. Cambridge Phil. Soc. 44, 391 (1948).
[8]C. Bloch, Phys. Rev. 93, 1094 (1954).
[9]A. A. Ross, Phys. Rev. 108, 720 (1957).

CAPTURE GAMMA RAYS FROM O^{15} AND O^{16} IN THE REGION OF THE GIANT RESONANCE*

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(Received October 15, 1959)

Measurements of the 90° yield ground-state γ rays resulting from the capture of protons into the giant resonance region of O^{15} and O^{16} have been using the Princeton FM cyclotron. This Letter reports principally the results of the reaction $N^{15}(p,\gamma)O^{16}$ ($Q = 12.11$ Mev).

The reaction $N^{15}(p,\gamma_0)O^{16}$ is the inverse of the reaction $O^{16}(\gamma,p_0)N^{15}$ where γ_0 and p_0 represent transitions to the ground state of the residual nucleus. The latter reaction has been studied by several authors[1-3] in the region of the giant resonance using the bremsstrahlung continuum as a source of high-energy photons. A detailed study of the giant resonance region of O^{16} is more readily made from the inverse process $N^{15}(p,\gamma_0)O^{16}$ using protons with a well-defined energy. The results of such a study are reported in this Letter for the region of excitation in O^{16} between 21 and 26 Mev. A detailed study of photodisintegration of O^{16} in this energy region has a particularly direct bearing on the understanding of the giant resonance phenomenon because the closing of the $1p$ shell at O^{16} allows a relatively explicit theoretical treatment to be made for this nucleus. Elliott and Flowers[4] have calculated the properties of the $J^{\pi} = 1^-$, $T=1$ states arising from the $p^{-1}d$ and $p^{-1}2s$ configurations in O^{16}. These states are responsible for electric dipole absorption from the O^{16} ground state and should give rise to the giant resonance according to the Wilkinson[5] model which explains the giant resonance in terms of single-particle excitations of the ground state. Elliott and Flowers found their calculations to be in substantial agreement with previous[1,2] investigations of the $O^{16}(\gamma,p)N^{15}$ reaction so that it is of interest to test their shell-model predictions more fully.

The experimental details will be given in a fuller report of this investigation; only a brief description is given here. The external proton beam from the Princeton FM cyclotron was reduced in energy with polyethylene absorbers to extend the range of variability of the proton beam energy from its unattenuated range of 14.5 to 19.5 Mev, so as to cover the range of the giant resonance, 9.5 to 15 Mev. The beam passed through a cylindrical gas cell $1\frac{1}{2}$ in. long, $\frac{3}{8}$ in. in diameter, which contained N^{15} gas at a pressure of 930 mm Hg. The end windows of the cell were of 0.0005-in. Mylar sheet. The N^{15} gas used has an isotopic purity of 98.7%. Gamma radiation from the target was detected in a 3 in. × 3 in. NaI crystal 7 cm from the center of the target, and the pulses were analyzed in a 200-channel analyzer gated on only for the duration of the cyclotron beam pulse. To minimize the effect of pulse pile-up in the counter, the current pulses from the photomultiplier were delay-line clipped to 0.2 μsec, and all pulses corresponding to an energy less than 10 Mev were eliminated at this stage by a biased transistor preamplifier. The larger pulses, passed by the biased preamplifier, were lengthened and fed to the analyzer.

Measurements were made at 29 proton energies. The beam current varied with energy, being about 3×10^{-5} μa at the lowest proton energy with the

thicker absorber (9.5 Mev) and about 2×10^{-4} μa at higher energies. Measurements were made for a constant integrated beam current of 0.75 microcoulomb.

The transition to the ground state stood out clearly at the highest energies. The next highest energy radiation present was that leading to the O^{16} excited states at ~6 Mev. In analyzing the data for the variation of cross section with energy, the counts contained within a range of 4.5 Mev of the maximum energy were included, and a background was subtracted on the assumption that the flat background observed at higher energies continued in the same way back into this region.

The variation of the 90° γ-ray yield with excitation energy in O^{16} is shown in Fig. 1. The differential cross section has been computed using a net efficiency for the NaI crystal of 0.25% at $E_\gamma = 22$ Mev. If it is assumed that the γ rays have an isotropic distribution relative to the proton beam, the ordinate scale of Fig. 1 gives the total cross section. The $N^{15}(p,\gamma_0)O^{16}$ results show two large resonances peaked at 21.8 and 24.7 Mev, respectively. There are strong indications of some detailed structure in the higher resonance. The peak energies are

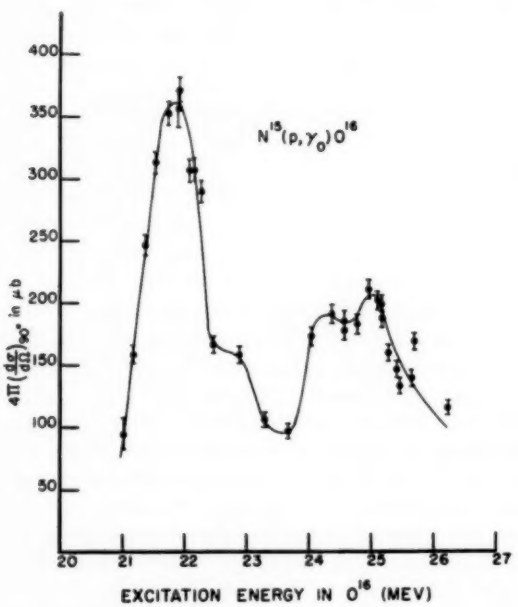

FIG. 1. The 90° yield of γ rays leading to the ground state of O^{16} from the $N^{15}(p,\gamma)O^{16}$ reaction. If the γ rays have an isotropic distribution relative to the proton beam, the ordinate scale gives the total cross section.

in striking agreement with the Elliott and Flowers calculations which predict $J^\pi = 1^-$, $T = 1$ levels at 22.6 and 25.2 Mev. Elliott and Flowers calculated the γ widths of the 22.6- and 25.2-Mev states to be in the ratio 1:2. From Fig. 1 it is seen that the present results are in qualitative agreement with this prediction, assuming that Γ/Γ_p is approximately the same for the two resonances, and would seem to give strong support for the single-particle model of the giant resonance, at least for the lighter nuclei.

The experimental arrangement used to investigate the $N^{14}(p,\gamma_0)O^{15}$ reaction ($Q = 7.30$ Mev) was similar to that described above except that a self-supporting melamine target of 4.9-mg/cm^2 thichness was used for the target; however, the results obtained are quite different from those obtained from $N^{15}+p$. For O^{15} excitation energies between 19 and 25 Mev the 90° yield of γ_0 was essentially constant with

$$4\pi\,(d\sigma/d\Omega)_{90°} \simeq 30\,\mu\text{b}.$$

This region of excitation in N^{15} (the mirror nucleus to O^{15}) has been studied by the $N^{15}(\gamma,p_0)C^{14}$ reaction.[6] The results indicate a narrow ($\Gamma \lesssim 500$ kev) resonance at ~20.5 Mev in N^{15} superimposed on a flat background. A possible reason why this resonance was not seen in the $N^{14}(p,\gamma_0)O^{15}$ reaction could be that it has $T = 3/2$, in which case it could not be formed by $N^{14}+p$.

We wish to acknowledge the warm encouragement of R. Sherr in this investigation. Two of the authors (S. G. C. and P. S. F.) are grateful to the State Department for Fulbright travel grants, and one of us (S. G. C.) wishes to express his appreciation for the hospitality extended by the Palmer Physical Laboratory during his sabbatical leave in 1959.

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‡Now at the Clarendon Laboratory, Oxford, England.

[1]Stephens, Mann, Patton, and Winhold, Phys. Rev. 98, 839 (1955).

[2]Sven A. E. Johansson and Bengt Forkman, Arkiv. Fysik 12, 359 (1957).

[3]P. Brix and E. K. Maschke, Z. Physik 155, 109 (1959).

[4]J. P. Elliott and B. H. Flowers, Proc. Roy. Soc. (London) A242, 57 (1957).

[5]D. H. Wilkinson, Physica 22, 1039 (1956).

[6]J. L. Rhodes and W. E. Stephens, Phys. Rev. 110, 1415 (1958).

MULTIPLE COULOMB EXCITATION IN Th^{232} AND U^{238}

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Electromagnetic excitation or "Coulomb excitation" of a nucleus by the electric field of a passing ion occurs most readily in nuclei having low-lying states with the character of collective excitations of the ground-state configurations.[1] Electric quadrupole excitations are by far the most important. "Multiple Coulomb excitation" refers to excitation of higher states through successive quadrupole excitation steps. In an earlier paper,[2] the double Coulomb excitation of even-even tungsten isotopes by O^{16} ions was reported (excitation of the 4+ member of the ground-state rotational band through the 2+ state).

The probability of multiple Coulomb excitation should increase rather rapidly with increasing projectile charge, and Coulomb excitation in general is most favorable where the rotational level spacings are smallest. With these factors in mind, search was made for higher than second-order Coulomb excitation in Th^{232} and U^{238} by irradiating with Ne^{20}, S^{32}, and A^{40} ions produced by the Berkeley heavy-ion linear accelerator (Hilac). These particular target elements also have the virtue of being monoisotopic. (The uranium used was isotopically depleted in U^{235} and U^{234}.) In this paper evidence is presented for excitation up to sixth order in U^{238} and fifth order in Th^{232}.

Because of low beam intensities (1-5 micro-microamperes) it was necessary to employ thick targets of thorium and uranium metal (2-4 mils). The gamma rays resulting from Coulomb excitation were detected with a 3 in. $\times$ 3 in. NaI(Tl) crystal assembly mounted behind the target and recorded with a 100-channel pulse-height discriminator. In order to enhance the events from multiple Coulomb excitation and to reduce the background radiation, the gamma-ray detector was operated in fast coincidence (3×10^{-8} sec) with pulses produced by the back-scattered heavy ions in an argon scintillation chamber. These gating pulses were cut off below about 10 Mev.

The gamma-ray spectra obtained with A^{40} irradiation of U^{238} and Th^{232} are shown in Figs. 1(a) and 1(b). The gamma-ray energies observed are listed in columns 2 and 5 of Table I except that the energies for the 2+ → 0+ transitions were taken from other experiments. The 4+ → 2+ transition in U^{238} (103 kev) is somewhat uncertain both in energy and intensity because uranium K x-rays lie at 94 to 98 kev and accurate resolution was not possible. In Figs. 1(a) and 1(b) the small peaks which appear beyond energies of 311 kev and 273 kev can be reasonably well explained both in energy and intensity, as due to coincident arrival at the detector of lower energy photons. Such pile-up peaks, occasioned by the high acceptance angle of the detector, would make a negligible contribution to the gamma rays listed in Table I.

The interpretation of these data in the form of level schemes is shown in Fig. 2. The evidence is largely indirect since it would be experimentally difficult to determine all of the gamma-gamma coincidences in the de-excitation sequences. However, as a partial check, it was established that in U^{238} the 214-kev (8+ → 6+)

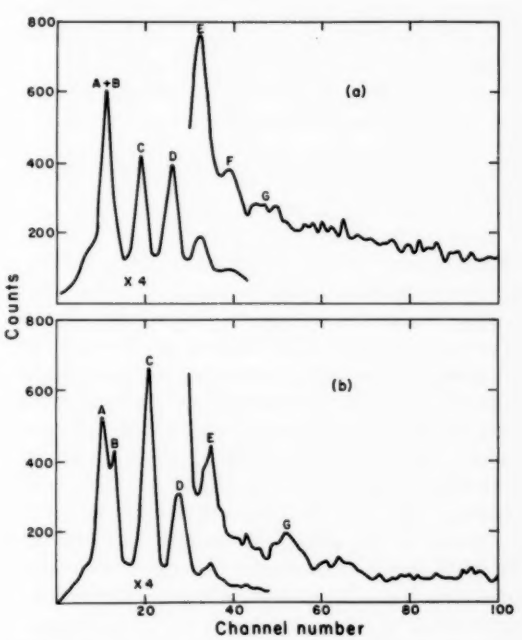

FIG. 1. De-excitation gamma-ray spectrum of (a) U^{238}, (b) Th^{232} irradiated with 190-Mev A^{40} ions. A, B, C, D, E, and F correspond to K x-rays, 4+ → 2+, 6+ → 4+, 8+ → 6+, 10+ → 8+, 12+ → 10+ transitions, respectively, and G is a pile-up peak (see text).

Table I. Transition energies between rotational states produced by multiple Coulomb excitation.

Transition	U^{238} transition energies (kev)			Th^{232} transition energies (kev)			
	Exper.	Calc. for $A=7.45$	Calc. for $A=7.46$, $B=-2.7\times10^{-3}$	Exper.	Calc. for $A=8.33$	Calc. for $A=8.38$, $B=-8.5\times10^{-3}$	Calc. for $A=8.34$, $B=-1.2\times10^{-2}$, $C=4\times10^{-5}$
2+→0+	(44.7 ± 0.2)[a]	44.7	44.7	(49.75 ± 0.25)[b]	50	50	50
4+→2+	103 ± 2	104	103	113 ± 2	117	114	113
6+→4+	161 ± 2	164	162	170 ± 3	183	173	170
8+→6+	214 ± 3	224	215	222 ± 4	250	222	221
10+→8+	264 ± 4	283	265	273 ± 5	317	259	272
12+→10+	311 ± 5	343	310				

[a]Taken from J. O. Newton, reference 5.
[b]This value was measured by the present authors from U^{236} α decay using a xenon-filled proportional counter.

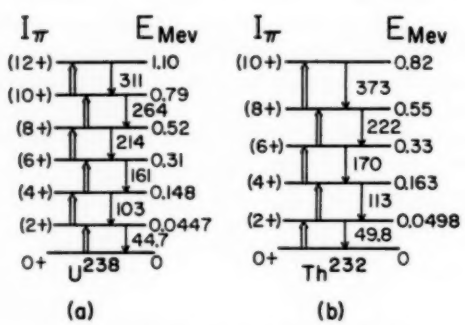

FIG. 2. Rotational energy level schemes proposed for (a) U^{238} and (b) Th^{232}.

gamma ray and part of the 103-kev (4+→2+) intensity are in coincidence with the 161-kev (6+→4+) transition. The principal pieces of evidence are the following: (1) The energies of the gamma rays are in agreement with expectations for excitation of rotational bands, (2) the yields of the photons for different bombarding ions and energies would be difficult to explain on any basis other than multiple Coulomb excitation of rotational bands, and (3) no other low-lying states are known in this region of the periodic system which would not at the same time give abundant crossover transitions of easily discernible energies.

It is now well known that levels of a rotational band of an even-even nucleus, such as those in Fig. 2, have energies given approximately by $E_I=AI(I+1)$, where E_I is the energy of the state with spin I and I is restricted to even integers.[3]

The empirically determined constant A is related to the effective moment of inertia, $\mathcal{I}$, by $A=\hbar^2/2\mathcal{I}$. The first excited states (2+) have been established by other Coulomb-excitation work,[4,5] and the energies obtained have been used to determine A. With this value of A the energies of other states may be calculated. The corresponding energy spacings of the rotational bands are listed in columns 3 and 6 of Table I. It is not surprising that agreement with the experimental results is increasingly poor for the higher states since it is expected (and found in other cases) that higher order terms should be added to the expression for E_I to account for rotational-vibration interactions. The second term $BI^2(I+1)^2$ contains another constant, B, which can be related to vibrational energies. Columns 4 and 7 of Table I show that this second term is sufficient to bring into agreement the U^{238} data; but for Th^{232} a third term, $CI^3(I+1)^3$, is required (column 8).

Table II gives a comparison of these constants for several nuclei in the heavy-element region.

Table II. Rotational constants in the heavy elements.

Nuclide	A (kev)	$-B$ (kev)	C (kev)
Pu^{240}	7.17	3.9×10^{-3}	
Pu^{238}	7.37	3.6×10^{-3}	
U^{238}	7.46	2.7×10^{-3}	$<2\times10^{-6}$
U^{234}	7.29	6.9×10^{-3}	$(\sim3\times10^{-5})$
Th^{232}	8.34	1.2×10^{-2}	4×10^{-5}
Ra^{226}	11.74	8×10^{-2}	$(\sim9\times10^{-4})$

As is well known, the value of A is rather constant for elements above thorium and increases rather sharply for lighter elements approaching the closed shells at Pb^{208}. Indeed, below radium the rotational picture is no longer a proper description of lowest-energy collective modes of motion. It is apparent that if accurate values for B and C are to be obtained and only a few members of a rotational band can be measured, great precision in the energy determination is required. However, if (as in the present study) a large number of states are discernible, high precision is not required. The values of C shown for Ra^{226} and U^{234} are only estimates which are entered to give some comparison with the more accurate values obtained in the present experiments.

Cross sections for Coulomb excitation have proved to be of great value because they are directly related to $E2$ transition probabilities and provide information for nuclear models. Such information on multiple Coulomb excitation would also be of interest as a guide in extending Coulomb excitation theory. Unfortunately, meaningful cross sections cannot be obtained from the present experiments because (1) it was necessary to use thick targets and the inherent difficulties in obtaining an excitation function are complicated by uncertainties in the dE/dx relation for argon ions; (2) only a portion of the excitations were observed, those for which the heavy ion was scattered through angles between 90° and 160° and emerged from the target with enough energy to enter the gas scintillation counter with greater than 5 to 10 Mev. Nevertheless some relative yields have been calculated for two different A^{40} energies (see Table III) and these will be explained presently.

There has been no published theoretical treatment applicable to high-order multiple excitation, but Alder and Winther[6] are now considering the problem. Their first results seem to be in excellent agreement with our observations in that qualitatively the predicted trends are being followed.

In columns 2 and 3 of Table III we have summarized the relative independent yields for excitation of the indicated levels in Th^{232} at two different A^{40} ion bombarding energies. These values were obtained from the integrated photopeaks corrected for absorption in the target and backing plate and for the counting efficiency of the NaI crystal. Correction was made for internal conversion (assuming $E2$ transitions), and allowance was made for the cascade from higher levels.

Table III. Relative yields from A^{40} on Th^{232}.

E_γ	Parent level	Direct yield of parent level $E(A^{40}) = 158$ Mev	$E(A^{40}) = 190$ Mev
113	4+	100	100
170	6+	46	88
222	8+	10	26
273	10+	0.7	5.9

The yield for the 4+ level has been normalized to 100 for each energy. It is seen that each successive level has an appreciably steeper excitation function so that at the higher bombarding energy the 10+ level is about eight times more intense relative to the 4+ than at the lower bombarding energy. This is explained in a natural way according to the scheme in Fig. 2, since here each higher state results from a higher order multiple excitation. Other explanations of this behavior are not so easy. If, for example, the gamma rays come from ordinary single $E2$ excitations from the ground state, then we would expect the 273-kev peak to increase only about 10% more than the 113-kev peak over this range of bombarding energy. To obtain the much greater variation in yield indicated in Table III, one would have to assume either a much higher (and different) energy of excitation for each transition, or excitations of higher multipole orders. Neither of these possibilities is very likely. The latter—the direct excitation of the rotational band members by $E4$, $E6$, etc., excitations instead of by the multiple process—was shown to be negligible in the case of double excitation in the tungsten isotopes using O^{16},[2] and should be even less important with a particle of higher charge. The yields seem entirely reasonable for the experimental conditions used (thick targets and back-scattered projectile ions). Had we used thin targets, the results of Winther and Alder would suggest that at the higher bombarding energy the primary population of the 6+ and perhaps the 8+ states would have been higher than for the 4+ state.[6] Such reversals in yield would be interesting to observe, but the difficulties associated with thin targets in our apparatus are rather severe. Data similar to those in Table III have been obtained for both U^{238} and Th^{232} with different bombarding ions. In all cases these data are consistent with the schemes in Fig. 2 insofar as it is possible to determine at the

present time.

We are grateful to Dr. J. O. Newton who participated in many of the early phases of this work. We are also greatly indebted to Dr. E. Hubbard and the operating crews of the Hilac for the indispensable help they have given us in carrying out these experiments.

[1]For a comprehensive review of the subject, see: Alder, Bohr, Huus, Mottelson, and Winther, Revs. Modern Phys. 28, 432 (1956).

[2]J. O. Newton and F. S. Stephens, Phys. Rev. Letters 1, 63 (1958).

[3]A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 27, No. 16 (1953).

[4]G. M. Temmer and N. P. Heydenburg, Phys. Rev. 93, 351 (1954); Davis, Divatia, Lind, and Moffat, Phys. Rev. 103, 1801 (1956); P. H. Stelson and F. K. McGowan, Phys. Rev. 99, 112, 616A (1955).

[5]N. P. Heydenburg and G. M. Temmer, Phys. Rev. 93, 906 (1954); 94, 1252 (1954); J. O. Newton, Nuclear Phys. 3, 345 (1951).

[6]K. Alder and A. Winther, Federal Institute of Technology, Zurich, Switzerland (private communication, June, 1959).

CAPTURE IN (K^-, p) ATOMS

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Day, Snow, and Sucher[1] have concluded that the reactions of K^- mesons at rest in liquid hydrogen almost invariably result from the interaction of the (K^-, p) system in states of zero relative orbital angular momentum. This conclusion results from a calculation, based on a suggestion by Madansky, that indicates that collisions of the protons in the liquid hydrogen with the (K^-, p) atom induce in the atom a strong Stark effect which mixes S-wave orbital states into states of higher orbital angular momentum. The very strong (K^-, p) S-wave absorption then depopulates these states before radiative transitions leading to the $2P$ state can occur. Previously, it had been generally believed that capture from the $2P$ state might be more important than radiative $2P \rightarrow 1S$ transitions, and that the (K^-, p) capture process might proceed predominantly through P-state capture. If it can be established that this view is incorrect and that capture almost invariably results from the S states, the parity of the K meson might be determined,[2] and other important conclusions might be reached, by a study of the $K^- - p$ and $K^- - d$ reactions for stopped K^- mesons. It is the purpose of this note to point out corrections to the specific calculations of Day et al. which appear to vitiate largely their specific conclusions that P-state capture cannot be important.

Day et al. consider the (K^-, p) atom moving essentially with thermal velocity through the liquid hydrogen. Frequent collisions with protons in the liquid result in the atom being subjected to intense electric fields which polarize the atom, or induce Stark transitions, for a period of time of the order of $2a_0/V$, where a_0 is the proton Bohr radius and V is the velocity of the atom. Dealing with the $n=6$ level, largely for the sake of definiteness, they calculate the Stark transition rate due to an electric field, $E = e/a_0^2$, of a proton at a distance of one Bohr radius. These transition rates are large, and Day et al. then conclude that the mixing is such that P states are completely depopulated in a collision while the other angular momentum states are rearranged into a $2l+1$ statistical population. They do not consider immediate capture from states of higher orbital angular momentum presumably because the collision time is not assuredly long compared to the time required to establish a complete mixing of states. Using these approximations, they follow an assembly of atoms through their history and conclude that, from statistically weighted $n=6$ states, 1.4% eventually reach the $2P$ level by radiative transitions while 98.6% are captured in S states. From states of higher n even fewer reach the $2P$ level.

The collisions considered by Day et al. are collisions of the second kind with a negligible energy transfer but with the transfer, from the (K^-, p) atom to the system of the atom and the colliding proton, of one or more units of angular momentum. The DeBroglie wavelength characteristic of an average collision is about 2π A while the effective collision radius a_0 used by Day et al. is a Bohr radius or about $\frac{1}{2}$ A. Since the ratio a_0/λ is then about $\frac{1}{2}$, classically collisions with $l \geq 1$ are forbidden. An estimate of the correction to

the results of reference 1 required by these considerations is easily made. The matrix element appropriate to a collision in which the (K,p) system in a state $\psi_{n,l}$ is changed to a state $\psi_{n,l-1}$, which can be the products of the $K-p$ interaction, is

$$\langle U_{L+1}{}^{*}\psi_{n,l-1}{}^{*}|H|U_{2}\psi_{n,l}\rangle,$$

where the U are the plane wave functions representing the relative (K,p) atom and proton coordinates and H, the Hamiltonian, will be equal to $(e^2/|R-r_p|)-(e^2/|R-r_k|)$, where R is the vector coordinate of the colliding proton and r_p and r_k are the coordinates of the proton and K meson in the atom. For $|R|>|r|$ a multipole expansion of H can be made. Setting $R=a_0$ as in reference 1, the matrix element can be rewritten as

$$\langle V_{L+1}{}^{*}(a_0)|V_{L}(a_0)\rangle\langle\psi_{n,l-1}{}^{*}|H'|\psi_{n,l}\rangle,$$

where, with appropriate averaging of geometric factors, the second term is precisely that evaluated by Day et al. In the first factor V represents the radial part of the wave functions U, and the square of this term has a value of 1/5 for S to P transitions which are the most favorable. Changes of more than one unit of angular momentum are much more strongly forbidden. These corrections modify the conclusions of reference 1 concerning the $n=6$ state in the following way. The depopulation of the P level in any collision is essentially unaffected but the reshuffling of other states is much reduced and their direct depopulation is largely forbidden. This greatly reduces the transfer into the P level and the average atomic lifetime is considerably increased, enhancing the importance of radiative transitions. Calculations of the same kind as reported in reference 1 lead then to the result that about 20% of atoms in a $n=6$ state reach the $2P$ state instead of the 1.4% stated in reference 1.

The uncertainties involved in the estimates made in this note, and also in reference 1, are quite large, and the conclusions reached in these calculations are not presented with the intention of establishing that P-wave capture is large, or that the Stark effect is unimportant. But we believe that these results do indicate the necessity of a more detailed examination of the problem.

[1]Day, Snow, and Sucher, Phys. Rev. Letters 3, 61 (1959).

[2]L. B. Okun' and I. A. Pomeranchuk, J. Exptl. Theoret. Phys. U.S.S.R. 34, 997 (1958) [translation: Soviet Phys. JETP 34, 688 (1958)].

GRAVITATIONAL RED-SHIFT IN NUCLEAR RESONANCE

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It is widely considered desirable to check experimentally the view that the frequencies of electromagnetic spectral lines are sensitive to the gravitational potential at the position of the emitting system. The several theories of relativity predict the frequency to be proportional to the gravitational potential. Experiments are proposed to observe the timekeeping of a "clock" based on an atomic or molecular transition, when held aloft in a rocket-launched satellite, relative to a similar one kept on the ground. The frequency ν_h and thus the timekeeping at height h is related to that at the earth's surface ν_0 according to

$$\Delta\nu_h=\nu_h-\nu_0=\nu_0 gh/c^2(1+h/R)$$
$$\approx\nu_0 h\times(1.09\times10^{-18}),$$

where R is the radius of the earth and h is the altitude measured in cm. Very high accuracy is required of the clocks even with the altitudes available with artificial satellites. Although several ways of obtaining the necessary frequency stability look promising, it would be simpler if a way could be found to do the experiment between fixed terrestrial points. In particular, if an accuracy could be obtained allowing the measurement of the shift between points differing as little as one to ten kilometers in altitude, the experiment could be performed between a mountain and a valley, in a mineshaft, or in a borehole.

Recently Mössbauer has discovered[1] a new aspect of the emission and scattering of γ rays by nuclei in solids. A certain fraction f of γ rays of the nuclei of a solid are emitted without

individual nuclear recoil. Instead, the recoil momentum is delivered to the crystal lattice as a whole resulting in negligible Doppler shift. Such γ rays are in resonance with nuclei similarly bound in a lattice and a similar fraction f of the electromagnetic resonant cross section

$$\sigma_R = 2\pi\lambda^2\left(\frac{2I_e+1}{2I_g+1}\right)\frac{1}{1+\alpha},$$

where I_e and I_g are the spins of the emitting and the ground states, respectively, and α is the internal conversion coefficient, pertains to the scattering. Calculations based on the Debye model of lattice vibrations yield for f at temperatures T much less than the Debye temperature θ_D

$$f = \exp\left\{-\tfrac{3}{2}\frac{E_\gamma^{\,2}}{2Mc^2k\theta_D}\left[1+\tfrac{2}{3}\left(\frac{\pi T}{\theta_D}\right)^2\right]\right\},$$

where E_γ is the energy of the γ ray, M is the nuclear mass, and k is Boltzmann's constant. The factor $(E_\gamma^{\,2}/2Mc^2k\theta_D)$ is the ratio of the recoil energy that would be taken up by the free nucleus to $k\theta_D$. For γ rays much above the 129 kev employed by Mössbauer the factor f becomes very small even at absolute zero.

The most striking evidence for the existence of this effect is the observation that the attenuation of the 129-kev gamma rays of Ir^{191} in passing through an iridium absorber is reduced if the source is moved. The speed required to reduce the part of the attenuation caused by resonant scattering to one-half its maximum value was found to be approximately 1.5 cm/sec. From this a half-life of the excited state is derived to be 0.1 mμsec. Others have repeated this experiment, and extended it to helium temperatures.[2,3] One other case is reported,[3] that of W^{182} wherein a half-life of 0.6 mμsec is inferred by the Doppler width of the resonance. This is half the accepted lifetime as measured by delay coincidence techniques. It is not clear whether this discrepancy represents a limit of the technique or whether it is largely an instrumental problem, as the authors suggest, enhanced by the complex array of other γ rays in the Ta^{182} source. Of course, as has been suggested, one should expect to see effects caused by hyperfine structure in these spectra when lifetimes are long enough to allow them to be important. All the effects discussed in connection with the directional correlation of cascade γ rays should have an influence. For example, it would seem desirable to use a source that has a good chance of being in a normal lattice site and electronic state at the time of emission of the final γ ray in question. One could have serious aftereffects from β decays, from prior emission of high-energy γ rays, or from electron captures as well as broadening from imperfections in the crystal lattice or short spin-lattice relaxation.

Even if the further development of the technique does not yield still narrower resonances, those already observed have fractional widths in frequency well below those of all the reference lines yet proposed for "atomic clocks." If the scattering is reduced to one-half its maximum by relative motion of the source and scatterer with velocity v, the Q, the ratio of the frequency to the full width at half-height of the resonance line being observed, is just $c/2v$. In the case of Mössbauer's experiment Q is about 1×10^{10} and in the case of W^{182} it is 7×10^{10}. In general $Q = 1.10E_\gamma(\text{Mev})\,\tau_{1/2}(\text{m}\mu\text{sec})\times10^{12}$.

A measurement of the gravitational red shift could be performed by transmitting γ rays from a source to a scatterer at an altitude different by h and by observing what relative velocity yields maximum scattering. For the predicted shift to be a full half-width of the line, the altitude difference h must be $h_{1/2} = [4.18/E_\gamma(\text{Mev})\,\tau_{1/2}(\text{m}\mu\text{sec})]$ km. Thus, for the width reported for W^{182}, 66 km difference of height would be required.

It is exciting to speculate about the possibilities opened up if cases of even less breadth can be found. For example, Fe^{57}, for which $E_\gamma = 0.0144$ Mev and $\tau_{1/2} = 100$ mμsec, would require only 2.9 km separation were it to yield its natural breadth. Another example might be Zn^{67} with an excited level at 0.093 Mev, of half-life 9400 mμsec. For this, if the natural breadth were obtained, $h_{1/2}$ would be 4.74 meters. This possibility represents a considerable extrapolation from present data. We are undertaking to examine these and other isotopes in various environments with the aim of selecting an isotope suitable for a gravitational experiment. Among other things equivalence of or absence of hyperfine structures in the sources and scatterers would be desirable.

Obviously one of the difficulties with large separations between source and scatterer arises from the inverse-square law of intensity. As a consequence of the participation of a large number of identical nuclei in an individual recoil-free scattering process, one anticipates the existence of intense Bragg diffraction from thin crystals. Thus one has the possibility of some

degree of focusing with bent crystals. Furthermore one may use the Bragg reflection from thin crystals to separate the γ rays emitted without recoil from all others. In this way irrelevant background γ rays could be eliminated from the detector.

Total external reflection of low-energy γ rays at grazing angles of incidence offers a possibility of a "light-pipe" to increase the effective solid angle that the scatterer subtends at the source. Within the limits set by the small angle of total reflection, this pipe need not be optically straight.[4]

The fixed baseline used for an experiment of this type reduces unwanted Doppler shifts to only those resulting from thermal, seismic, or similar disturbances. To equal the predicted gravitational shift the fractional change required in the height difference is 3.27×10^{-8} per second. Perturbing effects must be kept well below this value but this is also true for the other methods of measuring the red shift. Relative motion could be separated from the red shift by simultaneous observations of beams traveling in both directions.

[1]R. L. Mössbauer, Z. Physik 151, 124 (1958); Naturwissenschaften 45, 538 (1958); Z. Naturforsch. 14a, 211 (1959).

[2]Craig, Dash, McGuire, Nagle, and Reiswig, Phys. Rev. Letters 3, 221 (1959).

[3]Lee, Meyer-Schutzmeister, Schiffer, and Vincent, Phys. Rev. Letters 3, 223 (1959).

[4]We wish to thank E. M. Purcell for this suggestion.

ERRATUM

DISLOCATION PINNING IN n-TYPE GERMANIUM. R. L. Cummerow and A. R. Cherry [Phys. Rev. Letters 3, 367 (1959)].

Because it was thought that the diffusion coefficient of As in Ge is much smaller than it is, the experimental results presented in this Letter were interpreted in terms of Mott pinning (pinning of dislocations dragged through a field of immobile impurity atoms). This interpretation is valid for samples deformed in a few seconds time at 500°C, but at the annealing temperatures used (600°C and above), the more familiar Cottrell pinning (mobile impurity atom migrating to a fixed dislocation) is operating.

ABSTRACTS

In this section are printed the abstracts of Articles that have been forwarded to The American Institute of Physics for publication in THE PHYSICAL REVIEW. In quoting information obtained from this section before the appearance of the corresponding Article, reference should be made to "Physical Review (to be published)" rather than to this Journal.

ON THE QUANTUM THEORY OF THE THIRD VIRIAL COEFFICIENT. A. Pais, Institute for Advanced Study, Princeton, New Jersey, and G. E. Uhlenbeck, H. M. Randall Laboratory of Physics, University of Michigan, Ann Arbor, Michigan (Received May 20, 1959).

The quantum theory of the third virial coefficient C is discussed. Three types of intermolecular pair forces must be distinguished. (1) No bound or low-lying two- and/or three-body states. The first four terms of the low-temperature expansion of C are obtained. They depend on the scattering length, the effective range, and a third length which cannot be inferred from scattering data. The limitations of the applicability of such expansions are discussed, both for He^4 and He^3, by means of a comparison of the corresponding expansion for the second virial coefficient B with detailed numerical results known for specific potentials. (2) Existence of a near zero energy level both for the two- and the three-body system. It is shown how in this case the actual potentials may be replaced by suitably matched boundary conditions on the two- and the three-body wave functions near the respective coordinate origins. It is first explained how the method applies to B. Then the leading term of C is explicitly determined. (3) Existence of strongly bound two- and three-body states. An approximate expression for C is given by treating the single atoms and the binary and ternary compounds as a system of three ideal gases in chemical equilibrium.

SPECTRAL AND ANGULAR DISTRIBUTION OF CYCLOTRON RADIATION EMITTED BY COLLIDING PARTICLES. Ludwig Oster, Laboratory of Marine Physics, Yale University, New Haven, Connecticut (Received June 3, 1959).

An attempt is made to describe the cyclotron radiation of colliding particles in close analogy to optical theory of line formation. If the velocities are small compared with the velocity of light and the magnetic fields are not too strong, simple expressions are derived for the frequency and angular distribution of the radiation in terms of the basic physical properties of the plasma, such as temperature, density, and collision parameters.

TRANSPORT PHENOMENA IN SLIGHTLY IONIZED GASES: LOW ELECTRIC FIELDS. Mahendra Singh Sodha, Physics Division, Armour Research Foundation, Technology Center, Chicago, Illinois (Received June 5, 1959).

Chapman and Cowling have obtained the velocity distribution function of electrons in a slightly ionized gas when an electric field E is applied, by assuming elastic collisions and validity of Lorentzian approximation. The author has expanded this distribution function, neglecting terms involving fourth and higher powers of field, and used this expansion to calculate various transport properties, when a magnetic field is applied. To simplify the mathematics, the relaxation time has been taken as a power function of electron velocity. Some approximations have also been discussed. It is seen that at low electric fields the transport properties can be expressed as linear functions of E^2.

EFFECTS OF PARTICLE-PARTICLE INTERACTION ON THE MOMENT OF INERTIA OF MANY-FERMION SYSTEMS. Ronald M. Rockmore, Brookhaven National Laboratory, Upton, New York (Received June 2, 1959).

It is shown independent of perturbation-theoretic considerations that the effect of particle-particle interaction on the moment of inertia of a many-fermion system moving under periodic boundary conditions vanishes when calculated on the "cranking" model of Inglis. This result is obtained by performing a unitary transformation on the equivalent Hamiltonian which reproduces the principal sequence of diagrams in every order. The close resemblance of the general problem to the soluble model problem of a rotating dense electron gas is exploited in the calculation. This work extends that of Amado and Brueckner who have only demonstrated that interaction effects cancel in lowest order. The nature of their approximations and the connection

to the problem of collective excitation are discussed.

EFFECT OF SIMULTANEOUS DOPPLER AND COLLISION BROADENING AND OF HYPERFINE STRUCTURE ON THE IMPRISONMENT OF RESONANCE RADIATION. P. J. Walsh, Westinghouse Electric Corporation, Research Department, Lamp Division, Bloomfield, New Jersey (Received June 1, 1959).

The transmission coefficient defined by Holstein for resonance radiation has been calculated when Doppler and collision broadening of the resonance line are present simultaneously. From this a simple formula is inferred for the imprisonment lifetime of the resonance radiation under this condition. The complication of the hyperfine structure is also taken into account and the results are found to give good agreement with experiments on mercury.

GROUND STATE ENERGY AND EXCITATION SPECTRUM OF A SYSTEM OF INTERACTING BOSONS. N. M. Hugenholtz and D. Pines, The Institute for Advanced Study, Princeton, New Jersey (Received May 4, 1959).

In this paper properties of a boson gas at zero temperature are investigated by means of field-theoretic methods. Difficulties arising from the depletion of the ground state are resolved in a simple way by the elimination of the zero-momentum state. The result of this procedure when applied to the calculation of the Green's functions of the system is identical to that of Beliaev. It is then shown generally that for a repulsive interaction the energy $E(k)$ of a phonon of momentum k, which is found as the pole of a 1-particle Green's function, approaches zero for zero momentum, which means that the phonon spectrum does not exhibit an energy gap.

The Green's function method is applied to the calculation of the properties of a low-density boson gas. The next order term beyond that calculated by Lee and Yang, and Beliaev for the ground-state energy is obtained and the general form of the series expansion is found to be

$$(E_0/\Omega) = \tfrac{1}{2}n^2 f_0[1 + a(nf_0^3)^{1/2} + b(nf_0^3)\ln(nf_0^3) + c(nf_0^3) + d(nf_0^3)^{3/2}\ln(nf_0^3) + \dots],$$

where n is the density and f_0 is the scattering length for the assumed two-body interaction between the bosons. The coefficients a and b are independent of the shape of the interaction, and are the only terms thus far calculated. The coefficient b is in agreement with the hard-sphere gas calculations of Wu and of Sawada.

A discussion is given of the intermediate-density calculation of Brueckner and Sawada, and certain possible improvements in the method of summing a selected set of higher order terms are proposed.

BUILDUP OF A DISCHARGE IN ARGON. M. Menes, Physics Department, Westinghouse Research Laboratories, Beulah Road, Churchill Borough, Pittsburgh, Pennsylvania (Received June 3, 1959).

Measurements were made of the rate of buildup of an electrical discharge in argon in the pressure range from 5 to 60 cm Hg. The results are interpreted on the basis of a secondary mechanism due to delayed photons. The photon delay times which fit the observed data are in the neighborhood of five microseconds over the range of pressures investigated. These photon delay times are compared with (a) calculated imprisonment time for resonance radiation, and (b) delay times for molecular radiation as observed by Colli. Considering the uncertainty in the calculations and the lack of knowledge about the energy distribution between the two main resonance lines, the imprisonment times are of the right magnitude to explain the observed buildup rates. The Colli-process delay times are in fair agreement with the data at the higher pressures; at the low pressures they are definitely too slow to explain the observed buildup rates.

DOUBLE SCATTERING OF ELECTRONS WITH A DIPOLE MOMENT. M. H. Zaidi, Laboratory of Nuclear Studies, Cornell University, Ithaca, New York (Received May 28, 1959).

The double scattering of electrons with a small intrinsic electric dipole moment is investigated theoretically for the case where magnetic and electric fields are present in the space between the two targets. It is shown by using density matrix techniques that the plane of polarization of the electron beam, after the first scattering, would be rotated by the magnetic and electric

fields just as predicted by regarding the electrons as classical spinning tops with magnetic moments and classical electric dipoles precessing about the directions of the magnetic and electric fields, respectively.

NUCLEAR SPIN-LATTICE RELAXATION IN METALS. A. G. Anderson and A. G. Redfield, International Business Machines Watson Laboratory at Columbia University, New York, New York (Received June 5, 1959).

The nuclear spin-lattice relaxation time, T_1, has been measured in the range of 1.1°K to 4.2°K for the metals lithium, sodium, aluminum, and copper. A combination of nuclear magnetic resonance at fixed frequency and adiabatic variation of the magnetic field was used to measure T_1 as a function of field between zero and one thousand gauss. At fields of between one hundred and one thousand gauss T_1 is independent of magnetic field and inversely proportional to temperature in agreement with theory. The experimental values of the relaxation time multiplied by absolute temperature in sec °K are 44 ± 2.0 for Li^7, 5.1 ± 0.3 for Na^{23}, 1.80 ± 0.05 for Al^{27}, and 1.27 ± 0.07 for Cu^{63}. These values are in good agreement with previous experimental data at room temperature and above. At fields comparable with the nuclear magnetic dipole-dipole fields, T_1 is a function of applied field. The theory of relaxation in low fields is presented in an elementary form. Qualitative agreement with theory is obtained for Al^{27} and Cu^{63}; detailed agreement is obtained for Li^7 and Na^{23}.

ELECTRONIC SURFACE STATES AND THE CLEANED GERMANIUM SURFACE. Paul Handler and William M. Portnoy,* University of Illinois, Urbana, Illinois (Received June 5, 1959).

The large p-type surface conductivity and field-induced surface conductivity of the cleaned germanium surface have been measured over the temperature range 77-300°K. It is found that the surface conductivity and the field-induced surface conductivity are almost independent of temperature, varying by a factor of two over the temperature range investigated. The effect of oxygen, atomic hydrogen, and water vapor on the surface conductivity has also been observed. A qualitative two-dimensional band model is presented which correlates most of the experimental results. In this model, which is somewhat similar to the three-dimensional band model of graphite, there is a two-dimensional surface state band at the surface which overlaps in energy a two-dimensional valence band just beneath the surface. The states which form the surface state band are assumed to be perturbed out of the conduction band. The filling of these surface states with electrons out of the valence band gives rise to the observed p-type conductivity associated with the cleaned germanium surface.

The states perturbed out of the conduction band are shown to be associated with the unfilled orbitals of the germanium surface atoms. A second system, where a similar two-dimensional matrix of unfilled orbitals is found, is in the dislocations associated with medium-angle grain boundaries. It is shown that the transport properties of these grain boundaries are similar in magnitude and temperature dependence to those observed for the clean germanium surface. The same model presented for the surface can be used to explain the transport properties of grain boundaries.

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RESISTANCE TRANSITIONS IN SUPERCONDUCTING TANTALUM. D. P. Seraphim and R. A. Connell, International Business Machines Research Laboratory, Poughkeepsie, New York (Received June 3, 1959).

A study has been made of the superconducting to normal transition in outgassed tantalum wires induced by the application of an external longitudinal magnetic field. It was found that in most cases the transition, as determined from resistance measurements, consisted of an almost discontinuous appearance of resistance, followed by a rather gradual rise in resistance until the full residual resistance was restored. Flux measurements showed that the sharp portion of the transition corresponded to the magnetic or "bulk" transition. The behavior of the broad portion of the transition was of the type commonly associated with superconducting "filaments." Although the "filamentary" properties were found to be extremely sensitive to localized contamination and the thermal history of a specimen, they did not appear to depend exclusively

on such inhomogeneities, and a systematic variation with residual resistance could be discerned. Plastic deformation altered the characteristics only in proportion to the corresponding increase in residual resistance. A small amount of supercooling which was independent of the "filamentary" phenomena was sometimes observed in the "bulk" transition.

AC IMPEDANCE MEASUREMENTS ON INSULATED CdS CRYSTALS. Hartmut Kallmann, Bernard Kramer,* and Grace Marmor Spruch, Department of Physics, New York University, Washington Square, New York, New York (Received June 4, 1959).

In order to study the induced conductivity in CdS crystals without charge injection at the electrodes, the crystals were insulated with Mylar and ac impedance measurements were made. The capacitance and resistance of the crystals were studied as functions of intensity and wavelength of excitation, and frequency and voltage of the ac field. The crystal-Mylar combination was found to behave in a manner similar to that of powdered samples in like experiments. With a model that treats the crystal as a capacitor shunted by a light-sensitive resistance, the capacitance was found to increase with intensity of excitation, and to decrease with voltage and frequency. The resistance of the crystal increased by factors of 4 or 5 as the intensity of excitation decreased by factors of 10. The limitations of the model are discussed, and conclusions drawn regarding whether trapped electrons or only conduction electrons contribute to the impedance.

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EFFECTS OF ELECTRON CORRELATION ON THE OPTICAL PROPERTIES OF METALS. P. A. Wolff, Bell Telephone Laboratories, Murray Hill, New Jersey (Received June 4, 1959).

Many-body perturbation theory is used to determine the effect of electron-electron correlation on the optical properties of metals. The formula for the current induced in such a system by an electromagnetic wave is written in such a form that it may be conveniently evaluated with diagrammatic techniques. Lowest order diagrams give the usual conductivity of a noninteracting electron gas. Diagrams of first order (in the Coulomb field) are typical of the random-phase approximation and do not appreciably influence the conductivity. In second order, processes outside the random-phase approximation occur, and have an important effect on the optical properties. A formula is derived for the contribution of such diagrams to the conductivity. It gives a vanishing correction for the case of the free-electron gas (where current, as well as momentum, is conserved in electron-electron collisions) but a finite value for real metals in which electron current and momentum are not proportional to one another. This formula is used to discuss the optical properties of the double (electron-hole) plasma that occurs in metals with overlapping bands. In this instance correlation causes a shift in the dielectric anomaly (defined as the point at which the real part of the dielectric constant is zero), and produces absorption through processes in which an electron and hole share the energy of an incoming photon.

SELF-ENERGY OF A HELICAL DISLOCATION. Roland de Wit,* Department of Physics, University of Illinois, Urbana, Illinois (Received June 1, 1959).

Kröner's energy expression is used in this theoretical calculation. The helical dislocation is assumed to have a uniform shape with the Burgers vector along its axis. The axial length of the helix is large compared to its radius and the radius is large compared to the dislocation "cross section," which is of the order of a Burgers vector. For a helix of many turns and arbitrary pitch an expansion in Fourier cosine series is used. The self-energy is found in terms of elementary functions and Kapteyn series of Bessel functions. In the limiting cases of a tightly wound helix (small pitch) and a nearly straight helix (large pitch) simple expressions result, which have a plausible physical explanation. For a tightly wound helix the dominant term represents the contribution from the cylindrical part of the helix, the first-order terms represent the influence of the size of the dislocation cross section, and the second-order terms represent the effect of the axial component of the helix. For the nearly straight helix the

dominant terms represent the contribution from the straight screw part and the second-order terms are taken to give the interaction between the turns of the helix. Finally the correction in the self-energy when a return loop is present is considered.

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THEORY OF SURFACE MODES OF VIBRATION IN TWO- AND THREE-DIMENSIONAL CRYSTAL LATTICES. Richard F. Wallis, United States Naval Research Laboratory, Washington, D. C. (Received May 26, 1959).

Theoretical expressions have been developed for the frequencies and displacements of the normal modes of vibration for two- and three-dimensional alternating diatomic lattices with free boundaries. Only square and cubic lattices are considered. Nearest-neighbor Hooke's law forces having both longitudinal and transverse components are assumed. The results have been obtained both by a perturbation method in which the ratio of the transverse and longitudinal force constants is treated as a small quantity and by a Green's function method. The use of the free boundary condition leads to the existence of surface modes of vibration in which the displacement amplitude is relatively large for a light atom on a boundary and decreases roughly exponentially toward the interior of the lattice. A band of surface mode frequencies lies in the "forbidden" gap between the acoustical and optical branches.

LAW OF CORRESPONDING STATES IN SUPERCONDUCTIVITY. James C. Swihart, Research Center, International Business Machines Corporation, Poughkeepsie, New York (Received June 12, 1959).

It is shown by means of a crude calculation that a refined version of the BCS theory of superconductivity in which the energy gap function $\mu(\nu)$ would not be approximated by a constant would give very nearly the same relation between H_0, the critical field at $T=0$; $2\mu(0)$, the energy gap; and $N(0)$, the density of states at the Fermi surface. Agreement between the theory and experiment is found for the cases of Al, Sn, Hg, and Pb although the experimental error is large for Hg. For the case of Pb, this is the first agreement between the theory and experiment with regard to the thermodynamic variables.

VIBRATIONAL SPECTRUM OF VANADIUM. D. N. Singh and W. A. Bowers, Physics Department, University of North Carolina, Chapel Hill, North Carolina (Received June 1, 1959).

A theoretical lattice vibration spectrum is calculated for vanadium, in order to compare with the spectrum measured by the inelastic incoherent scattering of neutrons. The calculated spectrum shows three peaks, the upper two of which occur at frequencies agreeing with the frequencies of the two peaks in the experimental curve. The third peak is not present in the measured spectrum. The maximum frequency of the calculated spectrum is lower by about 15% than the measured value.

SLOW ELECTRONS IN POLAR CRYSTALS: SELF-ENERGY, MASS, AND MOBILITY. T. D. Schultz,* Department of Mathematical Physics, The University, Birmingham, England (Received September 30, 1959; revised manuscript received July 9, 1959).

The parameters for the Feynman model of a polaron are evaluated numerically for various values of the electron lattice interaction α, in the usual idealization of the problem of a slow electron in a polar crystal. The self-energy and effective mass thus obtained are compared with earlier polaron theories, indicating the superiority of the Feynman model for a wide range of α. The polaron size and the effect of the continuum approximation are estimated, and it is concluded that the alkali halides, at least, may be in the border region for the validity of this approximation. The problem of calculating polaron mobility as determined by scattering with longitudinal optical-mode phonons is analyzed and previous theories are critically reviewed. A new theory based on the Feynman model is developed in which the Boltzmann equation is used with resonance scattering considered as the fundamental scattering process. A comparison with previous theories shows some improvements and stresses still doubtful points. A comparison

with various experiments suggests the possible inadequacy of the usual idealization.

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ENERGY LEVELS IN NEUTRON-IRRADIATED *n*-TYPE SILICON. G. Rupprecht and C. A. Klein, Research Division, Raytheon Company, Waltham, Massachusetts (Received June 1, 1959).

Pulsed-field-effect experiments have been performed on neutron-irradiated *n*-type silicon samples with the aim of detecting deep-lying radiation-induced energy levels and estimating their electron capture cross sections. These experiments provide evidence in favor of two, and only two, deep-lying states in the upper part of the energy gap, at 0.15 ev and 0.37 ev below the conduction band edge. The electron capture cross sections associated with these two levels strongly suggest that both are acceptor-like.

SPATIAL DIFFUSION OF SPIN ENERGY. A. G. Redfield, International Business Machines Watson Laboratory at Columbia University, New York, New York (Received June 5, 1959).

A procedure is outlined for evaluating the spatial diffusion coefficient of magnetization of spins on a rigid lattice. The temporal recession of a spatially sinusoidally varying magnetization is analyzed, and is reduced to the problem of finding a function whose moments are known. An unambiguous value of diffusion coefficient can be obtained but the possibility of a complete lack of diffusion cannot be ruled out.

OPTICAL ABSORPTION OF CoO AND MnO ABOVE AND BELOW THE NÉEL TEMPERATURE. G. W. Pratt, Jr., Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts, and Roland Coelho, Laboratory for Insulation Research, Massachusetts Institute of Technology, Cambridge, Massachusetts (Received May 25, 1959).

The optical absorption of single crystal CoO and MnO both above and below the Néel temperature is reported and interpreted. The absorption spectrum in each case is similar to that found in dilute paramagnetic salts of the same ions. Tanabe and Sugano's strong-field theory was used to interpret the data. In CoO the tetragonal distortion of the cubic lattice present in the antiferromagnetic phase was experimentally observed. An estimate of the strength of the tetragonal field is given. It is also found that the temperature dependence of the absorption coefficient in CoO is in accord with a second-order electric dipole-phonon transition mechanism.

HIGH-FIELD EFFECT IN BORON-DOPED SILICON. R. D. Larrabee, RCA Laboratories, Princeton, New Jersey (Received May 18, 1959).

Two samples of boron-doped silicon were observed to have a linear current-voltage characteristic at liquid nitrogen temperature (77°K) up to fields of 10^4 volts/cm. This result was not expected since saturation of drift velocity is expected to occur at these high fields. Indeed, the current-voltage characteristic at 183°K did show the saturation effects expected. The experimental data seem to indicate that there is an appreciable amount (94%) of de-ionization of the boron level at 77°K and that the capture cross section of the ionized boron levels decreases as the hole drift velocity increases in the applied field. Since the thermal ionization rate is substantially independent of field, this implies that the steady-state number of carriers will be increased at the higher fields.

NEUTRON STUDY OF THE CRYSTAL AND MAGNETIC STRUCTURES OF $MnFe_{2-t}Cr_tO_4$. Stanley J. Pickart, U. S. Naval Ordnance Laboratory, White Oak, Silver Spring, Maryland, Brookhaven National Laboratory, Upton, New York, and University of Maryland, College Park, Maryland, and Robert Nathans, The Pennsylvania State University, University Park, Pennsylvania, and Brookhaven National Laboratory, Upton, New York (Received May 25, 1959).

Powder neutron diffraction measurements have been performed at temperatures down to 4.2°K on three compositions in the mixed spinel series $MnFe_{2-t}Cr_tO_4$ ($0 \leq t \leq 2$). Separation of the nuclear and magnetic scattering was accomplished both by applying an external field and by measuring the neutron intensities above the Curie

temperature. Analysis of the nuclear scattering indicates that all the compounds investigated have essentially the normal spinel structure, the fraction of divalent atoms on the tetrahedral sites being about 0.8, 0.9, and 1.0 for $t=0.5$, 1.0, and 1.5, respectively, while in every case the Cr atoms are located on the octahedral sites. The magnetic scattering is consistent with antiparallel A- and B-site moments, with no evidence being found for ordered triangular configurations. The magnitude of the A-site moments in each instance is approximately the moment value of the cations located there, while the B-site moments are significantly lower than expected on this basis. These results can be attributed to the occurrence within the B sublattice of spin-quenched cations, random antiparallel spins, or short-range ordered triangular configurations, since any of these alternatives lowers the sublattice moment on the average.

OPTICAL ANISOTROPY OF TETRAGONAL BARIUM TITANATE. Wataru Kinase, Department of Physics of General Education, Waseda University, Shinjuku, Tokyo, Japan, and Jinzo Kobayashi and Noboru Yamada, Department of Applied Physics, Waseda University, Shinjuku, Tokyo, Japan (Received May 18, 1959).

The optical dielectric constants and the anisotropy of tetragonal barium titanate were calculated by assuming spherical symmetry for the electron clouds of its constituent atoms. An analogous calculation was carried out for lead titanate. The calculated birefringences are positive in both crystals, in contrast to the observed ones. The results are discussed.

FREQUENCY DEPENDENCE OF THE SURFACE RESISTANCE OF SUPERCONDUCTING TIN IN THE MILLIMETER WAVELENGTH REGION. R. Kaplan,* A. H. Nethercot, Jr.,† and H. A. Boorse,‡ Columbia Radiation Laboratory, Columbia University, New York, New York (Received December 12, 1958; revised manuscript received June 26, 1959).

The ratio of the superconducting to normal surface resistance of polycrystalline tin has been measured at seven frequencies between 17 kMc/sec and 77 kMc/sec and at temperatures from 1.5°K to 3.0°K. These data plus those of other investigators have been compared with the predictions of two theories: the first a calculation made by Serber (unpublished) based on the London two-fluid model of superconductivity and the Reuter-Sondheimer theory of the anomalous skin effect, and the second a calculation based on the Bardeen-Cooper-Schrieffer theory as developed by Bardeen and Mattis.

Agreement between experimental and theoretical results is only fair in the case of the two-fluid theory. The best values of the relevant parameters, Fermi velocity v and mean free path l, were found to be, respectively, $(1.25 \pm 0.3) \times 10^7$ cm/sec and 10^{-3} - 10^{-1} cm. A value of v of approximately 10^8 cm/sec would be expected for tin. The surface resistance ratio from the Bardeen-Cooper-Schrieffer theory has been calculated only for the extreme anomalous limit and the calculation therefore should not apply too accurately for tin. However, a curve of the right general shape is obtained and further calculations more appropriate to tin should improve the agreement between theory and experiment.

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THEORY OF FERROMAGNETIC RESONANCE IN RARE EARTH GARNETS. II. LINE WIDTHS. P.-G. de Gennes, C. Kittel, and A. M. Portis, Department of Physics, University of California, Berkeley, California (Received June 8, 1959).

The spins of rare earth ions in the garnets are coupled strongly both to the lattice phonons and, by an exchange interaction, to the ferric spin lattice. The rare earth spins thus provide a powerful relaxation channel for the ferric lattice. Two contributions to the line width may be distinguished: a coherent process (in which the total magnetic moment of the ferric lattice relaxes without changing the magnitude of the moment) is dominant at temperatures from 0°K up to just below the Curie temperature. Near and above T_c a fluctuation process (in which ferric spins flip locally) is dominant. The theoretical results describe the order of magnitude and the temperature dependence of the observed line widths in the rare earth garnets and in impure

yttrium iron garnet, if one assumes in the absence of direct experimental knowledge that the relaxation frequency $1/\tau$ of the relevant rare earth ions is $\sim 10^{-12}$ sec at 400°K. As the temperature is increased from 0°K, the width increases until a maximum is reached when $1/\tau$ becomes comparable with the ferric-rare earth exchange frequency. Above this temperature the width decreases until near T_c, where there is a sharp rise.

FORMATION OF INTERSTITIALS IN ALKALI HALIDES BY IONIZING RADIATION. R. E. Howard and R. Smoluchowski, Carnegie Institute of Technology, Pittsburgh, Pennsylvania (Received June 8, 1959).

Recent experimental evidence seems to indicate that interstitials in alkali halides are formed by ionizing radiation. The validity of the so-called Varley mechanism depends on several factors such as the lifetime of the positive halogen ion, lattice geometry, etc. These are evaluated and found favorable.

TIME DELAYS IN THE SUPERCONDUCTING TRANSITION OF LEAD FILMS. R. F. Broom and E. H. Rhoderick, Services Electronics Research Laboratory, Baldock, Herts, England (Received May 18, 1959).

Strips of lead between 500 A and 1000 A thick, evaporated onto mica substrates, were driven from the superconducting into the normal state by rectangular current pulses of 0.4-μsec duration. For current amplitudes just above the threshold value, there was an apparent delay of up to 0.4 μsec before resistance began to appear in the strip. A plausible explanation is that a minute portion of the strip in the neighborhood of a flaw is driven normal almost instantaneously, and that the Joule heating of this normal region eventually causes thermal propagation of the interphase boundary. The delay is the time that must elapse before the temperature of the nucleus rises sufficiently to initiate the thermal spreading process. Similar results were obtained with lead-indium alloys, but in the case of tin the delay was less than the instrumental resolution. The relevance of these results to the interpretation of dc critical currents is discussed.

CURRENT TRANSITIONS IN SUPERCONDUCTIVE TIN FILMS. J. W. Bremer and V. L. Newhouse, General Electric Company, Schenectady, New York (Received May 25, 1959).

Except at temperatures just below the critical temperature, the change of resistance of a superconducting film if the current through it is increased slowly takes place by a discontinuous transition, exhibits hysteresis, and is dominated by Joule heating effects. By applying current in short pulses, it has been possible to obtain isothermal transitions in films deposited on a flat surface. These transitions are smooth, do not exhibit hysteresis, and have a shape independent of film resistance. The critical current I in films of thickness about twice the penetration depth is proportional to film width and can be related to the critical field of the film H_C, and the bulk critical field H_{CB}, by the relation $IH_C = \text{const}\, H_{CB}^2$. It is also shown that by calculating the conditions of thermal equilibrium the dc transition curves with Joule heating can be derived from the isothermal transition results.

DEVIATIONS FROM MATTHIESSEN'S RULE IN ALUMINUM, TIN, AND COPPER ALLOYS. P. Alley* and B. Serin, Department of Physics, Rutgers University, New Brunswick, New Jersey (Received May 18, 1959).

The electrical resistance of aluminum, tin, and copper alloys was measured at 4.2°K, 77°K and from 198°K to 348°K. The aluminum alloys contained zinc, magnesium, germanium, or silver in various concentrations. The alloys of tin contained indium, antimony, or bismuth; and those of copper contained zinc. At temperatures above 77°K, the impurity resistivity, $\delta\rho$, for a given solvent may be described by the equation $\delta\rho = \alpha(T)\rho_\gamma$, where ρ_γ is the residual resistivity, measured at 4.2°K, and T is the temperature. This result is independent of the nature of the solute. α is constant in the temperature region 198°K to 348°K. For aluminum and tin $\alpha(273) = 1.12$, whereas for copper $\alpha(273) = 1.05$. The value of $\alpha(77)$ is about the same as at 273°K for aluminum and copper, but for tin $\alpha(77) = 1.08$. The temperature coefficient of the impurity resistivity for $198°K < T < 348°K$ was no larger than 1×10^{-3} $(°K)^{-1}$. It is pointed out that while several different theoretical models qualitatively describe these results, none can be quantitatively

compared with experiment.

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NEW PARALLEL PHOTOELECTROMAGNETIC EFFECT. A. Amith, Radio Corporation of America Laboratories, Princeton, New Jersey (Received May 21, 1959; revised manuscript received July 27, 1959).

A new photoelectromagnetic effect is described, in which the short-circuit current is proportional to the difference in surface recombination velocities of a pair of parallel surfaces. The surfaces are the yz planes of a slab which is illuminated in the z direction in the presence of a magnetic field in the same orientation. In addition to the primary diffusion of the generated carriers along z there ensues a secondary flow in the transverse x direction. If the boundary conditions at the two yz planes are not identical, there results a net flow of carrier pairs toward one of these surfaces. The magnetic field B_z deflects these carriers, and a net short-circuit current passes in the y direction. Measurement of this current affords a means of probing the surfaces from the interior of the sample. The theory of the effect and its experimental observation are described.

LIFETIME OF THE 2S STATE OF ATOMIC HYDROGEN. Wade L. Fite, R. T. Brackmann, David G. Hummer, and R. F. Stebbings, John Jay Hopkins Laboratory for Pure and Applied Science, General Atomic Division of General Dynamics Corporation, San Diego, California (Received May 25, 1959).

A sensitive test for any mixing of the $2S_{1/2}$ and $2P_{1/2}$ state of atomic hydrogen is the measurement of the rate for single-quantum decay of the 2S atom to the ground state. A new upper limit of this decay rate has been determined. A section along a beam of 2S atoms, produced by electron excitation of a ground-state atom beam, was viewed by an iodine-vapor-filled ultraviolet photon counter, which responds to the Lyman-alpha radiation of the single-quantum decay process. From the counts observed when an electrostatic quenching field was superposed on the counter's field of view, the necessary experimental parameter (product of 2S atom current and counter efficiency) was determined. With the field removed, a portion of the remaining counts could be ascribed to quenching on collision of the 2S atoms with residual gases in the vacuum chamber, the quenching cross sections for which were measured. The decay rate not ascribable to known quenching effects was 420 sec^{-1}. Since unknown quenching effects may have been operative, this figure must be considered only as an upper limit for the natural single-quantum decay rate.

COLLISIONS OF ELECTRONS WITH HYDROGEN ATOMS. IV. EXCITATION OF LYMAN-ALPHA RADIATION NEAR THRESHOLD. Wade L. Fite, R. F. Stebbings, and R. T. Brackmann, John Jay Hopkins Laboratory for Pure and Applied Science, General Atomic Division of General Dynamics Corporation, San Diego, California (Received May 25, 1959).

Using improved experimental techniques, the cross section for excitation of Lyman-alpha radiation in collisions between electrons and hydrogen atoms has been remeasured. It has been determined that in the threshold region, the results reported previously were somewhat low. The remeasured energy dependence of the cross section near threshold has been found to be as the square root of the excess energy.

THEORY OF THE SCATTERING OF ELECTRONS FROM ATOMIC HYDROGEN. A. Temkin, National Bureau of Standards, Washington, D.C. (Received May 26, 1959).

The distortion of atomic hydrogen by a slowly moving electron at a large distance from the center of the atom is examined. The problem is the initial phase of a previously described method for the calculation of electron scattering which takes this distortion into account. The initial (static) problem is solved analytically and extended to include higher order effects of the interaction of the electron with the atomic cloud. The construction of a wave function to describe scattering starting from the solution of the static problem is clarified. This yields as an incidental result a new approximation of the second order perturbation energy associated with the above distortion. A short discussion of the present

experimental results for this scattering process is included.

POWER SATURATION OF THE $J = 1 - 2$ ROTATIONAL TRANSITION OF OCS. A. Dymanus,* Fysisch Laboratorium der Rijksuniversiteit, Utrecht, Netherlands (Received May 25, 1959).

The power saturation of seven lines arising from the $J = 1 - 2$ rotational transition in OCS has been investigated with a Stark-cavity spectrometer. The experimental results on all investigated lines confirm the theory of Karplus and Schwinger adapted in this paper to the case of gas enclosed in a cylindrical cavity absorption cell oscillating in a TE_{0mn} mode with an arbitrary n-value, and an m-value from 1 to 10.

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HALF-LIFE AND BETA SPECTRUM OF Rb^{87}. K. F. Flynn and L. E. Glendenin, Argonne National Laboratory, Lemont, Illinois (Received June 1, 1959).

Liquid scintillation counting techniques have been applied to the measurement of the specific activity and the beta spectrum of the natural radioactivity of rubidium. The measured Rb^{87} half-life is $(47.0 \pm 1.0) \times 10^9$ yr, and the observed maximum beta energy is 272 ± 3 kev.

ELECTRON CAPTURE DECAY OF Lu^{173} TO LEVELS IN Yb^{173}. J. W. Bichard and J. W. Mihelich, University of Notre Dame, and B. Harmatz, Oak Ridge National Laboratory (Received June 3, 1959).

The electron capture of Lu^{173}(1.30-year) to levels in Yb^{173} has been investigated using scintillation spectrometers and permanent magnet spectrographs. Levels in Yb^{173} were observed at 78.7 (7/2-), 179.5 (9/2-), 351.2 (7/2+), and 636.8 (7/2-) kev. The first two excited states may be interpreted as members of a rotational band based on the ground state [(512) 5/2-]. The levels at 351 and 637 kev are probably the intrinsic levels [(633) 7/2+] and [(514) 7/2-], respectively, predicted by Nilsson. L electron capture was observed to be the primary mode of population of the 637-kev level. Relative reduced transition probabilities were obtained for all transitions to the ground-state rotational band, and comparisons with theoretical branching ratios were made. Logft values were estimated from the observed K-capture branching ratios.

NUCLEAR RESONANCE FLUORESCENCE IN Ce^{140}. S. Ofer* and A. Schwarzschild, Brookhaven National Laboratory, Upton, New York (Received June 5, 1959).

Measurements of the lifetime of the 1.6-Mev first excited state of Ce^{140} have been performed using the nuclear resonance fluorescence method. Self-absorption experiments using Ce metal and CeO_2 scatterers and absorbers performed at 300°K and at 78°K yield a mean life of $(1.10 \pm 0.15) \times 10^{-13}$ second for this state. Formulas are presented for the analysis of resonance fluorescence experiments using thick scatterers and absorbers.

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NUCLEAR REACTIONS IN STARS. IV. BUILDUP FROM CARBON. H. Reeves* and E. E. Salpeter, Laboratory of Nuclear Studies, Cornell University, Ithaca, New York (Received June 1, 1959).

Reactions leading to the formation of C, O, and Ne at temperatures T of about 10^8 °K, in the core of red giant stars, had been studied previously. The nuclear reactions and resulting nucleogenesis are now studied for a gas consisting of C^{12}, O^{16}, and Ne^{20} at 6×10^8 to 10×10^8 °K and densities around 10^4 g/cc. The basic reaction rates are calculated and a set of simultaneous differential equations for various abundances as a function of time is solved numerically.

The C^{12} is destroyed by compound nucleus formation from C + C collisions in about 10^5 years and 1 year, respectively, at 6×10^8 and 8.5×10^8 °K. The net result is the production of some additional amounts of O^{16} and Ne^{20}, appreciable amounts of Na^{23} and the three stable magnesium isotopes (mainly Mg^{24}), and decreasing amounts of Al^{27}, Si, etc. The ratio of Na^{23} to Mg^{24} produced is almost unity, appreciably larger than the "cosmic" ratio.

During the carbon burning, protons and alpha particles are released. For $T \lesssim 7 \times 10^8$ °K, neutrons are produced very copiously through the

sequence $C^{12}(p,\gamma)N^{13}$; $N^{13}-C^{13}$; $C^{13}(\alpha,n)O^{16}$. If small amounts of metals in the Fe region were present in the gas originally (abundance x by number of C^{12} nuclei) they are transformed into heavy elements. For $x=10^{-3}$, about 90 neutrons are absorbed per Fe nucleus (more neutrons still for smaller values of x). For $T \geq 8\times10^{8}$ °K, the neutron production is appreciably less since N^{13} is photodisintegrated.

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MEASUREMENT OF THE L/K-CAPTURE RATIO IN Fe^{55} DECAY. J. Scobie, R. B. Moler, and R. W. Fink,* Department of Chemistry, University of Arkansas, Fayetteville, Arkansas (Received June 4, 1959).

The L and K x-radiations of manganese resulting from orbital-electron capture in a gaseous source of Fe^{55} have been studied in a multiwire proportional counter. The L/K-capture ratio was found to be 0.108 ± 0.006, in good agreement with the theoretical value.

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LIFETIMES OF 2^+ ROTATIONAL STATES. M. Birk, G. Goldring, and Y. Wolfson, Weizmann Institute of Science, Rehovoth, Israel (Received June 4, 1959).

The mean lives of the rotational 2^+ states of nuclei in the region $150 \leq A \leq 186$ have been measured with a pulsed proton beam timing device. The presence of prompt proton bremsstrahlung requires more elaborate analysis of the time distributions than the customary determination of center-of-gravity shift. In order to achieve the necessary accuracy and freedom from drift, a beam switching procedure was introduced, alternating the beam between two targets and therefore allowing accurate comparisons to be made. A measurement of the mean life of B^{10}, carried out as a check, yielded results in good agreement with previous measurements. A method for utilizing the lifetime measurements for accurate determinations of $B(E2)$ values is discussed.

SURFACE COUPLING MECHANISM FOR APPROACHING STATISTICAL EQUILIBRIUM IN COMPOUND NUCLEUS FORMATION, WITH APPLICATION TO FISSION. Lawrence Wilets, Physics Department, University of Washington, Seattle, Washington (Received May 27, 1959).

In a particular representation where the state vectors are not eigenstates of the Hamiltonian, coupling terms remain which cause "virtual" or "real" transitions among states. An appropriate choice of representation depends upon the physical processes involved. The decay of a compound nucleus, especially by fission, is considered. The strong coupling representation of the unified model is employed, with surface oscillations inducing transitions among states of the representation. A diffusion equation is derived to describe the flow of probability among the states available within constraints. An estimate of the characteristic relaxation time for arriving at statistical equilibrium is obtained. Only when the relaxation time is short compared with a basic reaction time are statistical arguments valid to evaluate the reaction rate. As an example, the relevant reaction time in the fission process is a collective vibrational period. The necessary condition appears to be satisfied for excitation energies more than a few Mev above threshold. Arguments are presented to show why equilibrium may not be maintained at lower energy. Thus the usual estimate of the number of open channels, $2\pi\bar{\Gamma}_f/D$, would give a number lower than what one would estimate simply from penetrability of the fission barrier. This seems to explain, at least in part, the anomalously low numbers of channels obtained in this manner. Problems relating to the validity of often made statistical assumptions at scission are also discussed.

SPALLATION-FISSION COMPETITION IN HEAVY-ELEMENT REACTIONS: $Th^{232}+He^4$ AND $U^{233}+d$. Bruce M. Foreman, Jr.,* Walter M. Gibson,† Richard A. Glass,‡ and Glenn T. Seaborg, Lawrence Radiation Laboratory and Department of Chemistry, University of California, Berkeley, California (Received May 18, 1959).

Cross sections and excitation functions have been determined for spallation and fission products from bombardments of Th^{232} with helium ions (15 to 46 Mev) and U^{233} with deuterons (9 to

24 Mev). This work extends a series of investigations of reactions induced by charged particles (α, d, and p) in heavy elements ($Z \geq 88$). Radiochemical methods were employed to isolate products corresponding to the following spallation reactions: neutron emission, $(\alpha, 4n)$, $(\alpha, 5n)$, (d, n), $(d, 2n)$, and $(d, 3n)$; emission of one proton and neutrons, (α, p), (α, pn), $(\alpha, p2n)$, and $(\alpha, p3n)$; and emission of two protons and neutrons, $(\alpha, 2p)$, $(\alpha, 2pn)$, $(\alpha, \alpha n)$, and $(d, \alpha n)$. In addition, the following fission products were isolated from one or more bombardments: Zn^{72}, Ge^{77}, As^{77}, $Br^{82,83}$, Rb^{86}, $Sr^{89,91}$, Y^{93}, $Zn^{95,97}$, Nb^{96}, Mo^{99}, $Ru^{103,105,106}$, $Pd^{109,112}$, Ag^{111}, $Cd^{115,115m,117}$, $I^{131,133}$, Cs^{136}, $Ba^{139,140}$, La^{140}, $Ce^{141,143,144}$, Nd^{147}, Eu^{157}, and Gd^{159}.

The results show that fission is the predominant reaction at all energies for Th^{232} and to an even greater extent for U^{233}. The data for the surviving spallation products are consistent with several mechanisms of reaction, including compound-nucleus formation and evaporation, direct interactions between nucleons of the incoming helium ion or deuteron and nucleons of the nucleus, and a combination of these types of processes (direct interaction followed by evaporation). In general, the results confirm and extend previously established concepts.

The neutron-emission spallation reactions as well as fission are best explained as proceeding through compound-nucleus formation. The shapes and magnitudes of $(\alpha, 4n)$, $(d, 2n)$, and $(d, 3n)$ excitation functions correlate well with a compound-nucleus treatment modified to include fission competition. According to this treatment, ratios of neutron to total-reaction level width, $\Gamma_n/\sum_i \Gamma_i$, are 0.49 for $U^{236-233}$ [from $Th^{232}(\alpha, 4n)$], 0.17 for $Np^{235-234}$ [from $U^{233}(\alpha, 2n)$], and 0.20 for $Np^{235-233}$ [from $U^{233}(d, 3n)$]. In addition the total-reaction excitation functions (consisting mostly of the fission excitation functions) are consistent with theoretical cross sections for compound-nucleus formation calculated with a nuclear radius parameter $r_0 = 1.5 \times 10^{-13} A^{1/3}$.

The fission mass-yield curves are similar to those found for other heavy target isotopes (for elements from thorium to plutonium). The minimum in the curves in the region of mass 120 tends to disappear as helium-ion or deuteron energy is increased.

The (α, pxn), $(\alpha, 2pxn)$, $(\alpha, \alpha n)$, (d, n), and $(d, \alpha n)$ products are attributed to direct interactions, with complex particles emitted in preference to a series of protons and neutrons. Thus (α, d), (α, t), and (α, tn) mechanisms would account for most of the (α, pn), $(\alpha, p2n)$, and $(\alpha, p3n)$ products, respectively. In the case of the (α, t) and (α, tn) reactions, analysis of the ratio $\sigma(\alpha, tn)/\sigma(\alpha, t)$ leads one to the conclusion that with 35-Mev helium ions only 9% of outgoing tritons leave the residual nucleus with sufficient energy to evaporate a neutron or undergo fission, and with 44-Mev helium ions only 20% do so. The (d, n) product probably results from the stripping reaction.

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PLAUSIBILITY OF A NONLOCAL OPTICAL MODEL. Y. C. Tang, R. H. Lemmer, P. J. Wyatt, and A. E. S. Green, Department of Physics, Florida State University, Tallahassee, Florida (Received May 18, 1959).

The implication of a simple two-kernel form of nonlocal potential is considered in the nuclear matter approximation. It is shown that this leads to a wave equation with a complex reduced mass. The parameters characterizing the real part of the optical potential are found to be in a reasonable accord with expectations from two-body forces. The parameters associated with the imaginary part are handled only phenomenologically. A description is found which works quite well in the energy range 0 to 25 Mev. The results of this study compares favorably with the corresponding results of an investigation with a nonlocal model for finite nuclei.

NUCLEAR SPIN, HYPERFINE-STRUCTURE SEPARATION, AND MAGNETIC MOMENT OF 22-HOUR POTASSIUM-43. F. Russell Petersen, Vernon J. Ehlers, W. Bruce Ewbank, Lawrence L. Marino, and Howard A. Shugart, Department of Physics and Lawrence Radiation Laboratory, University of California, Berkeley, California (Received June 1, 1959).

With the atomic-beam magnetic-resonance method, the nuclear spin and hyperfine-structure separation have been measured for 22-hour K^{43}.

The results are $I=3/2$, $\Delta\nu(^2S_{1/2})=192.64\pm0.05$ Mc/sec. The nuclear magnetic moment calculated from these measurements is $|\mu|=0.163\pm0.002$ nuclear magneton.

EFFECT OF NUCLEAR FORCES ON THE CROSS SECTIONS OF PHOTONUCLEAR REACTIONS. K. Okamoto,* Department of Physics, Louisiana State University, Baton Rouge, Louisiana (Received May 20, 1959).

The effect of nuclear forces, or the effect of the quasi-deuteron model, is discussed for the integrated cross section and the bremsstrahlung weighted cross section. The nuclear force is assumed to be of partly Majorana exchange character. Only the central force is considered. The two-body potential is a Gaussian type without a hard core, the parameters of which are taken from the effective-range theory. The calculations are performed by first order perturbation theory and the results are that the integrated cross section is increased by about ten percent and the bremsstrahlung weighted cross section is decreased by a few percent. Therefore the independent-particle model can be regarded as a good approximation for photonuclear reactions.

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MASS ASSIGNMENTS AND SOME DECAY CHARACTERISTICS OF Gd^{145}, Eu^{145}, Gd^{146}, AND Eu^{146}. J. Robb Grover, Chemistry Department, Brookhaven National Laboratory, Upton, New York (Received May 21, 1959).

The nuclides Gd^{145}, Eu^{145}, Gd^{146}, and Eu^{146} were prepared by the interactions of 20- to 40-Mev helium ions with Sm^{144}, and found to decay with half-lives of 25 ± 2 minutes, 5.6 ± 0.3 days, 46 ± 2 days, and 4.4 ± 0.1 days, respectively. The mass number assignments were made on the basis of excitation functions, and chemical evidence of parent-daughter relationships, with special reference to the previously known nuclide Eu^{145}. The most prominent gamma rays appearing in the decay of each of these four nuclides are as follows: in Gd^{145} decay, at 0.80, 1.03, and 1.75 Mev; in Eu^{145} decay, at 0.53, 0.64, and 0.89 Mev; in Gd^{146} decay, at 0.114 and 0.153 Mev; and in Eu^{146} decay, at 0.63 and 0.74 Mev. There is also a strong K x-ray line in each spectrum. In addition, Gd^{145} was found to emit positrons with an end-point energy of about 2.4 ± 0.2 Mev.

MATHEMATICAL ANALYSIS OF A SIMPLE MODEL RELATED TO THE STRIPPING REACTION. Elliott Lieb* and Heinz Koppe,† Department of Physics, University of Illinois, Urbana, Illinois (Received May 20, 1959).

A scattering process in which an incoming bound particle can split up into its component parts when its total energy is above a threshold is of considerable interest physically (deuteron stripping, etc.) and mathematically (analytic properties of the S-matrix, etc.). For such complicated problems it is obviously convenient to have a simple, analytically soluble model for reference, but although many models have been suggested in the past none have proven analytically tractable. In this paper we propose and completely solve a one-dimensional model which, although it is not very physical, has all the desired characteristics. The problem is not mathematically trivial however, and leads to a Wiener-Hopf integral equation.

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IMAGINARY PART OF THE OPTICAL POTENTIAL. L. C. Gomes,* Department of Physics and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts (Received May 25, 1959).

The imaginary part of the optical potential has been investigated for low-energy incoming neutrons, by means of the nucleon-nucleon cross sections in nuclear matter. The cross sections have been calculated under the assumption that pair correlations for low excited states of nuclear matter are the same as those formed in the ground state. The dependence of the effective mass on the single-particle momentum has been taken into consideration using an empirical solution which reproduces the present assumptions for the single-particle spectrum. The results have been applied to the nuclear surface in the Thomas-Fermi approximation. The maximum in the imaginary potential was found to be at the sur-

face outside of the half-density radius. For low incident energies it is about 1.5×10^{-13} cm beyond this radius.

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HYPERFINE STRUCTURE AND NUCLEAR MOMENTS OF BROMINE-82. Hugh L. Garvin, Thomas M. Green, Edgar Lipworth, and William A. Nierenberg, Lawrence Radiation Laboratory, University of California, Berkeley, California (Received May 20, 1959).

The nuclear spin, the magnetic dipole, and the electric quadrupole interaction constants have been measured for 35-hr bromine-82 by the method of atomic beams. The results are $I=5$, $|a|=205.04\pm0.05$ Mc/sec, $|b|=-870.7\pm0.9$ Mc/sec, and $b/a=-4.246\pm0.001$.

The nuclear magnetic and electric quadrupole moments obtained from these values of a and b are $|\mu|=1.6264\pm0.0005$ nuclear magnetons, and $|Q|=0.76\pm0.03$ barn. While only the relative sign of μ and Q is determined, both μ and Q are almost certainly positive. A new method for solving the interaction Hamiltonian with magnetic field, for arbitrary I and J, by using an IBM-650 computer, is described.

MODIFIED HARTREE-FOCK METHOD FOR THE FINITE NUCLEUS. K. A. Brueckner and D. T. Goldman, Department of Physics, University of Pennsylvania, Philadelphia, Pennsylvania (Received June 1, 1959).

A procedure is given for inclusion of the rearrangement energy term in the Brueckner-Gammel method for evaluating the structure of finite nuclei. A variational technique is used to derive the single-particle energy from the total binding energy of the nucleus.

It is seen that although a rearrangement energy term does not appear explicitly in the total nuclear binding energy, it is derivable from the binding energy and influences the eigenfunctions of the total nuclear system, and thus its energy eigenvalues. The rearrangement energy has noticeable effect on the single-particle wave equation and must be included in finite-nucleus calculations. To a good approximation, it is shown that it is possible to evaluate the rearrangement potential and to express it as a simple function of the mean nuclear density, i.e., $V_R = 240$ Mev $\rho^2(\times10^{-13}$ cm$)^6$.

COMPARISON OF (p,p') REACTIONS WITH (d,d') AND (p,n) REACTIONS. Bernard L. Cohen, University of Pittsburgh, Pittsburgh, Pennsylvania (Received May 18, 1959; revised manuscript received June 15, 1959).

It is pointed out that experimental results indicate very striking similarities between (p,p') and (d,d') reactions, and very striking differences between (p,p') and (p,n) reactions. It is shown that this is in strong disagreement with the nucleon-nucleon collision model commonly used in interpreting (p,p') reactions, but in agreement with the recent inelastic diffraction scattering model of Blair.

REACTIONS OF ALPHA PARTICLES WITH GERMANIUM-70 AND ZINC-70. Saadia Amiel,* Chemistry Department, Brookhaven National Laboratory, Upton, New York (Received May 21, 1959).

The reactions $Ge^{70}(\alpha,2n)Se^{72}$, $Ge^{70}(\alpha,pn)As^{72}$, $Zn^{70}(\alpha,pn)Ga^{72}$, and $Zn^{70}(\alpha,2p)Zn^{72}$ were studied with alpha particles of 20-40 Mev, and their excitation functions were measured. The results are compared with evaporation calculations based on the assumption of compound-nucleus formation.

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π^+-p INTERACTIONS AT 500 Mev. William J. Willis,* Yale University, New Haven, Connecticut, and Brookhaven National Laboratory, Upton, New York (Received May 6, 1959).

The interaction of π^+ mesons with protons at an energy of 500 Mev has been studied in a hydrogen bubble chamber. Phase shift analyses with S and P waves were made, and a near degeneracy was found between the Fermi and Yang solutions. When D waves were included, an additional ambiguity was found. The D-wave phase shifts are small, but they have a considerable effect on the other phase shifts. The cross section for single pion production is 2.85 ± 0.5 mb. The ratio

$(p+0)/(n+)$ is $1.5^{+1.5}_{-0.5}$. The cross section leading to $p++-$ was found to be of the order of 30 μb.

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SCATTERING OF 3.7- TO 25-Mev POSITIVE PIONS BY HYDROGEN. G. E. Fischer* and E. W. Jenkins,† Department of Physics, Columbia University, New York, New York (Received June 10, 1959).

The Columbia University hydrogen bubble chamber was used to investigate the π^+-p scattering cross section in a laboratory energy range from 3.7 to 25 Mev. A total of 950 events were measured, of which 338 were caused by incident pions that would have come to rest in the chamber. Treating the small p-wave and large Coulomb contributions as known, the s-wave phase shift is found to deviate from a linear dependence on momentum only by one and a half standard deviations.

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HEAVY NUCLEI IN THE PRIMARY COSMIC RADIATION AT PRINCE ALBERT, CANADA. PART I: CARBON, NITROGEN, AND OXYGEN. H. Aizu,* Y. Fujimoto, S. Hasegawa, M. Koshiba, I. Mito,† J. Nishimura, and K. Yokoi,† Institute for Nuclear Study, University of Tokyo, Tanashi, Tokyo, Japan, and Marcel Schein, Enrico Fermi Institute for Nuclear Studies, Department of Physics, University of Chicago, Chicago, Illinois (Received May 22, 1959).

A stack of G-5 emulsion, exposed at 120 000 feet for 8 hours at 61°N, has been used to study the charge and energy spectrum of heavy nuclei at the low-energy end. Energy measurements have been made on C, N, and O nuclei up to 1 Bev/nucleon. The spectrum shows a broad maximum at 550 Mev/nucleon, extrapolated to the top of the atmosphere. Various possibilities to explain this spectrum are discussed. However, it seems desirable to determine the energy spectrum of the other heavy nuclei components in this energy region in order to gain a more complete understanding of the whole problem. Measurements of this kind are in progress and will be reported.

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ENERGY DETERMINATION FROM MULTIPLE MESON PRODUCTION BY 6.3-Bev PROTONS IN NUCLEAR EMULSION. P. L. Jain and H. C. Glahe, Department of Physics, University of Buffalo, Buffalo, New York (Received May 28, 1959; revised manuscript received July 29, 1959).

864 stars produced by the 6.3-Bev proton beam of the Berkeley Bevatron were observed in nuclear emulsion. The average number of shower particles was 2.7 prongs per star. The energy of the primary particle was determined by three different methods: (a) the median angle method; (b) Castagnoli's method; (c) Cocconi's graphical method. It is found that the results of these three methods are in good agreement with each other and are high in each case by a factor of 2.

SUDDEN INCREASE OF COSMIC-RAY INTENSITY. Hugh R. Anderson, Norman Bridge Laboratory of Physics, California Institute of Technology, Pasadena, California (Received June 15, 1959).

A sudden 30% increase in cosmic ray intensity lasting approximately 12 minutes was observed at an atmospheric depth equal to 80 g/cm^2 by a Neher integrating ionization chamber flown from Bismarck, North Dakota, on October 16, 1958. A similar measurement made simultaneously at Invercargill, New Zealand, observed no increase. These observations are not in accord with the simple solar impact zone theory.

HIGH-ENERGY BEHAVIOR IN QUANTUM FIELD THEORY. Steven Weinberg, Department of Physics, Columbia University, New York, New York (Received May 21, 1959).

An attack is made on the problem of determining the asymptotic behavior at high energies and momenta of the Green's functions of quantum field theory, using new mathematical methods from the theory of real variables. We define a

class A_n of functions of n real variables, whose asymptotic behavior may be specified in a certain manner by means of certain "asymptotic coefficients." The Feynman integrands of perturbation theory (with energies taken imaginary) belong to such classes. We then prove that if certain conditions on the asymptotic coefficients are satisfied, then an integral over k of the variables converges, and belongs to the class A_{n-k} with new asymptotic coefficients simply related to the old ones. When applied to perturbation theory, this theorem validates the renormalization procedure of Dyson and Salam, proving that the renormalized integrals actually do always converge, and provides a simple rule for calculating the asymptotic behavior of any Green's function to any order of perturbation theory.

ANALYTIC PROPERTIES OF PARTIAL AMPLITUDES IN MESON-NUCLEON SCATTERING. S. W. MacDowell,* Mathematical Physics Department, University of Birmingham, Birmingham, England (Received January 26, 1959).

The analytic properties of partial wave amplitudes in meson-nucleon scattering are investigated on the basis of the Mandelstam's representation and an integral representation is set up for them which explicitly exhibits those properties.

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MULTIPOLE SINGULARITIES OF CLASSICAL VECTOR AND PSEUDOVECTOR FIELDS. Peter Havas, Department of Physics, Lehigh University, Bethlehem, Pennsylvania (Received May 15, 1959).

The general form of the equations of motion of a particle possessing multipole singularities of a neutral vector or pseudovector meson field has been found by Harish-Chandra under the assumption of a divergenceless current. In a recent paper a more general form of the interaction was proposed, and the equations of motion were found for this case for neutral, charged, and charge-symmetric fields. In this paper we establish forms of the multipole moments of arbitrary order compatible with these equations, and also of the moments compatible with the more restrictive interactions proposed by Harish-Chandra, which include those of electrodynamics as a special case. They are established under the assumption that the spin of the particle is of constant magnitude and has only spatial components in the rest system, and for pure 2^n-pole moments of constant magnitude ("intrinsic moments") or of variable magnitude ("induced moments," acceleration-dependent forces). In the Appendix the results of a previous paper on singularities of scalar and pseudoscalar fields, which established the general form of the intrinsic moments, are generalized to show the possibility of induced moments of all orders, and it is shown that a particle can carry an arbitrary linear combination of multipole singularities of spin-zero and spin-one fields.

ELECTROMAGNETIC CORRECTIONS TO ISOTOPIC SPIN CONSERVATION. S. Weinberg,* Columbia University, New York, New York, and S. B. Treiman, Palmer Physical Laboratory, Princeton University, Princeton, New Jersey (Received May 25, 1959).

If electromagnetic interactions are wholly responsible for all departures from isotopic spin invariance, then the strict conservation law $\Delta T = 0$ may be replaced, to order e^2, by the rule $|\Delta T| \leq 2$. Consequences of this weaker restriction are discussed for elementary particle masses, scattering processes, and weak-interaction decay processes. The apparent absence in nature of particles with isotopic spins greater than one makes it difficult to find very practical experimental tests of this rule.

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GREEN'S FUNCTIONS FOR ELEMENTARY PARTICLES. Hiroshi Ezawa and Hiroomi Umezawa, Department of Physics, University of Tokyo, Tokyo, Japan (Received May 18, 1959).

Recently, Harris has raised an objection against our work on the general structure of the Green's function for particles of arbitrary spin. Reasons are given here why his objection is not valid.